

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
10 January 2002 (10.01.2002)

PCT

(10) International Publication Number  
**WO 02/02527 A1**

(51) International Patent Classification<sup>7</sup>: C07D 213/61,  
213/64, 213/80, 213/70, 213/74, 213/26, A01N 43/40

(21) International Application Number: PCT/JP01/05851

(22) International Filing Date: 5 July 2001 (05.07.2001)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:  
2000-203909 5 July 2000 (05.07.2000) JP  
2001-034182 9 February 2001 (09.02.2001) JP  
2001-094222 28 March 2001 (28.03.2001) JP

(71) Applicant (for all designated States except US): ISHIIHARA SANGYO KAISHA, LTD. [JP/JP]; 3-15, Edobori 1-chome, Nishi-ku, Osaka-shi, Osaka 550-0002 (JP).

(72) Inventors; and

(75) Inventors/Applicants (for US only): NISHIDE, Hisaya [JP/JP]; c/o Ishihara Sangyo Kaisha, Ltd., Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525-0025 (JP). OGAWA, Munekazu [JP/JP]; c/o Ishihara Sangyo Kaisha, Ltd., Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525-0025 (JP). KOMINAMI, Hidemasa [JP/JP]; c/o Ishihara Sangyo Kaisha, Ltd., Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525-0025 (JP). HIGUCHI, Koji [JP/JP]; c/o Ishihara Sangyo Kaisha, Ltd., Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome,

Kusatsu-shi, Shiga 525-0025 (JP). NISHIMURA, Akihiro [JP/JP]; c/o Ishihara Sangyo Kaisha, Ltd., Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525-0025 (JP).

(74) Agents: SENMYO, Kenji et al.; Torimoto Kogyo Bldg., 38, Kanda-Higashimatsushitacho, Chiyoda-ku, Tokyo 101-0042 (JP).

(81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

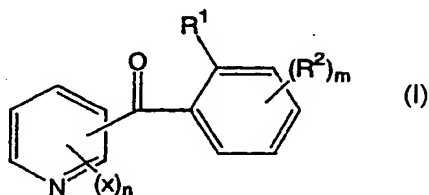
(84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

**Published:**

- with international search report
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: BENZOYL-PYRIDINE DERIVATIVE OR ITS SALT, FUNGICIDE CONTAINING IT AS AN ACTIVE INGREDIENT, ITS PRODUCTION PROCESS AND INTERMEDIATE FOR PRODUCING IT



(57) Abstract: The present invention relates to a fungicide containing a novel benzoylpyridine derivative or its salt. The present invention provides a fungicide containing a benzoylpyridine derivative represented by formula (I) or its salt: wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is a substitutable alkyl group; R<sup>2</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group; and m is 1, 2, 3 or 4, provided that when m is at least 2, R<sup>2</sup> may contain an oxygen atom to form a condensed ring.

**THIS PAGE BLANK (USPTO)**

DESCRIPTION

BENZOYLPYRIDINE DERIVATIVE OR ITS SALT, FUNGICIDE  
CONTAINING IT AS AN ACTIVE INGREDIENT, ITS PRODUCTION  
PROCESS AND INTERMEDIATE FOR PRODUCING IT

5 TECHNICAL FIELD

The present invention relates to a novel  
benzoylpyridine derivative or its salt, a fungicide  
containing it as an active ingredient, its production  
process and an intermediate for producing it.

10 BACKGROUND ART

Benzoylpyridine derivatives which are analogous to  
the compounds of the present invention may be compounds  
as disclosed in e.g. WO99/41237, WO99/38845, WO96/17829,  
JP-A-7-309837 and JP-A-2-275858. However, they are  
15 different from the compounds of the present invention.  
Further, the purposes of use of these compounds are  
different from those of the compounds of the present  
invention.

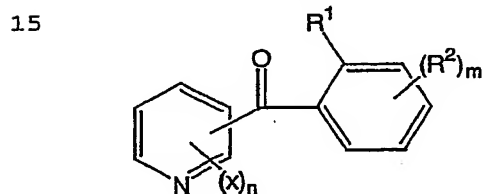
Many fungicides which have been conventionally  
20 provided have their own characteristics in their  
controlling effects over pests which cause plant  
diseases. Some have a slightly poorer curative effect as  
compared with a preventive effect, and some have a  
residual effect which lasts only for a relatively short  
25 period of time, so that their controlling effects against  
pests tend to be practically insufficient in some cases.  
Accordingly, it has been desired to develop a novel

compound which has a strong controlling effect against pests which cause plant diseases.

# DISCLOSURE OF THE INVENTION

The present inventors have conducted extensive studies to overcome the above problems and as a result, have found that use of the compound represented by the formula (I) as an active ingredient presents excellent preventive effect and curative effect against various plant diseases, particularly powdery mildew of barley, vegetables, fruits and flowering plants, and the present invention has been accomplished.

Namely, the present invention relates to a benzoylpyridine derivative represented by the formula (I) or its salt:



wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R¹ is a substitutable alkyl group; R² is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a

substitutable cycloalkoxy group or a hydroxyl group; and  
m is 1, 2, 3 or 4, provided that when m is at least 2, R<sup>2</sup>  
may contain an oxygen atom to form a condensed ring  
(excluding a case where the pyridine ring is substituted  
5 by a benzoyl group at the 2-position; the pyridine ring  
is substituted by an alkoxy group, a hydroxyl group or a  
benzyloxy group at the 3-position; and n is 1, m is 1 or  
2), a fungicide containing it as an active ingredient,  
its production process and an intermediate for producing  
10 it.

The halogen atom represented by X may, for example,  
be fluorine, chlorine, bromine or iodine, and preferably  
fluorine, chlorine or bromine, may, for example, be used.

The alkoxy moiety in the substitutable alkoxy group  
15 represented by each of X and R<sup>2</sup> may, for example, be a  
C<sub>1-6</sub> alkoxy (such as methoxy, ethoxy, propoxy, isopropoxy,  
butoxy, isobutoxy or t-butoxy), and preferably it may,  
for example, be a C<sub>1-4</sub> alkoxy (such as methoxy or ethoxy).  
Further, the secondary substituent of the substitutable  
20 alkoxy group may be from one to five substituents which  
are the same or different, selected from the group  
consisting of an aryl, an aryloxy, hydroxyl, nitro,  
nitroxy, a halogen (such as fluorine, chlorine, bromine  
or iodine), a haloalkoxy (such as a C<sub>1-4</sub> haloalkoxy such  
25 as CF<sub>3</sub>O or HCF<sub>2</sub>O), a cycloalkyl, amino, an alkylthio and  
cyano. Of these substitutable alkoxy groups, preferred  
is an alkoxy group which is not substituted, particularly

preferred is a C<sub>1-4</sub> alkoxy group.

As the aryl moiety in the substitutable aryloxy group represented by X, a condensation type polycyclic group such as naphthyl as well as phenyl may be  
5 mentioned, and preferred is phenyl. The secondary substituent of the substitutable aryloxy group may, for example, be a halogen, an alkyl, an alkoxy or hydroxyl. Of these substitutable aryloxy groups, most preferred is a phenoxy group.

10 The cycloalkyl moiety in the substitutable cycloalkoxy group represented by X is usually one having a carbon number of from 3 to 10, and a monocyclic group such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl or cyclooctyl, as well as a condensation type polycyclic  
15 group, may, for example, be mentioned. However, preferred is a monocyclic group. The secondary substituent of the substitutable cycloalkoxy group may, for example, be a halogen, an alkyl, an alkoxy or hydroxyl. Of these substitutable cycloalkoxy groups,  
20 most preferred is a cyclohexyloxy group.

The hydrocarbon moiety in the substitutable hydrocarbon group represented by X may, for example, be a C<sub>1-6</sub> alkyl group (such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl or t-butyl), a C<sub>2-6</sub> alkenyl  
25 (such as vinyl, allyl, isopropenyl or 3-methyl-2-butenyl), a C<sub>2-6</sub> alkynyl (such as ethynyl, 1-propynyl or 2-propynyl), a C<sub>3-6</sub> cycloalkyl (such as cyclopropyl,

cyclopentyl or cyclohexyl), or a C<sub>6-10</sub> aryl. Further, the secondary substituent of the substitutable hydrocarbon group may be from one to five substituents which are the same or different, selected from the group consisting of an aryl, an aryloxy, hydroxyl, nitro, nitroxy, a halogen (such as fluorine, chlorine, bromine or iodine), a haloalkoxy (such as a C<sub>1-4</sub> haloalkoxy such as CF<sub>3</sub>O or HCF<sub>2</sub>O), a cycloalkyl, amino, an alkylthio and cyano. Of these substitutable hydrocarbon groups, preferred is a substitutable alkyl group, and particularly preferred is an alkyl group. Further, a C<sub>1-4</sub> alkyl group is most preferred among alkyl groups.

The alkylthio moiety in the substitutable alkylthio group represented by X may, for example, be a C<sub>1-6</sub> alkylthio (such as methylthio, ethylthio, propylthio, isopropylthio, butylthio, isobutylthio or t-butylthio), and preferably a C<sub>1-4</sub> alkylthio (such as methylthio or ethylthio) may, for example, be mentioned. Of these alkylthio groups which may be substituted, preferred is an alkylthio group, particularly preferred is a C<sub>1-4</sub> alkylthio group. The secondary substituent of the substitutable alkylthio group may be from one to five substituents which are the same or different, selected from the group consisting of an aryl, an aryloxy, hydroxyl, nitro, nitroxy, a halogen (such as fluorine, chlorine, bromine or iodine), a haloalkoxy (such as a C<sub>1-4</sub> haloalkoxy such as CF<sub>3</sub>O or HCF<sub>2</sub>O) and cyano.

The carboxyl group which may be esterified or amidated, represented by X, may, for example, be a carboxyl group which may be esterified such as a C<sub>1-6</sub> alkoxy carbonyl group (such as a methoxy carbonyl group, an ethoxy carbonyl group, a propoxy carbonyl group, an isopropoxy carbonyl group, a butoxy carbonyl group, an isobutoxy carbonyl group or t-butoxy carbonyl group), a nitroxy C<sub>1-4</sub> alkoxy aminocarbonyl group (such as 2-nitroxy ethoxy carbonyl group or a 3-nitroxy propoxy carbonyl group), a phenyl C<sub>1-4</sub> alkoxy carbonyl group (such as a benzyloxy carbonyl group or a phenethyloxy carbonyl group); or a carboxyl group which may be amidated such as a carbamoyl group, a C<sub>1-6</sub> monoalkyl aminocarbonyl group (such as a methyl aminocarbonyl group, an ethyl aminocarbonyl group, a propyl aminocarbonyl group, an isopropyl aminocarbonyl group, a butyl aminocarbonyl group, an isobutyl aminocarbonyl group or a t-butyl aminocarbonyl group), a C<sub>1-6</sub> dialkyl aminocarbonyl group (such as a dimethyl aminocarbonyl group, a diethyl aminocarbonyl group, a dipropyl aminocarbonyl group, a diisopropyl aminocarbonyl group, a dibutyl aminocarbonyl group or an isobutyl aminocarbonyl group), a nitroxy C<sub>1-4</sub> alkyl aminocarbonyl group (such as a 2-nitroxy ethyl aminocarbonyl group or a 3-nitroxy propyl aminocarbonyl group), a phenyl C<sub>1-4</sub> alkyl aminocarbonyl group (such as a benzyl aminocarbonyl group or a phenethyl aminocarbonyl group), a C<sub>3-6</sub>

cycloalkylaminocarbonyl group (such as a cyclopropylaminocarbonyl group, a cyclopentylaminocarbonyl group or a cyclohexylaminocarbonyl group), a cyclic aminocarbonyl group (such as a morpholinocarbonyl group, a piperidinocarbonyl group, a pyrrolidinocarbonyl group or a thiomorpholinocarbonyl group) or an aminocarbonyl group.

The substitutable amino group represented by X may, for example, be an amino group or an alkylamino group such as a monoalkylamino group or a dialkylamino group. The alkyl moiety in the alkylamino group (a monoalkylamino group or a dialkylamino group) is preferably a C<sub>1-4</sub> alkyl. The secondary substituent of the substitutable amino group may be from one to five substituents which are the same or different, selected from the group consisting of an aryl, an aryloxy, hydroxyl, nitro, nitroxy, a halogen (such as fluorine, chlorine, bromine or iodine), a haloalkoxy (such as a C<sub>1-4</sub> haloalkoxy group such as CF<sub>3</sub>O or HCF<sub>2</sub>O), a cycloalkyl, amino, an alkylthio and cyano.

The alkyl moiety in the substitutable alkyl group represented by each of R<sup>1</sup> and R<sup>2</sup> is preferably a C<sub>1-6</sub> alkyl (such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl or t-butyl), and among them, preferred is a C<sub>1-4</sub> alkyl. The secondary substituent of the substitutable alkyl group may be from one to five substituents which

are the same or different, selected from the group consisting of an aryl, an aryloxy, hydroxyl, nitro, nitroxy, a halogen (such as fluorine, chlorine, bromine or iodine), a haloalkoxy (such as a C<sub>1-4</sub> haloalkoxy such as CF<sub>3</sub>O or HCF<sub>2</sub>O), a cycloalkyl, amino, an alkylthio and cyano. Of these substitutable alkyl groups, preferred is an alkyl group which is not substituted, and particularly preferred is a C<sub>1-4</sub> alkyl group. Among them, most preferred is a methyl group.

10       The alkoxy moiety in the substitutable alkoxy group represented by R<sup>2</sup> is preferably a C<sub>1-6</sub> (alkoxy such as methoxy, ethoxy, propoxy, isopropoxy, butoxy, isobutoxy or t-butoxy), and among them, preferred is a C<sub>1-4</sub> alkoxy. The secondary substituent of the substitutable alkoxy  
15       group may be from one to five substituents which are the same or different, selected from the group consisting of an aryl group, an aryloxy group, a hydroxyl group, a nitro group, a nitroxy group, a halogen atom (such as fluorine, chlorine, bromine or iodine), a haloalkoxy  
20       group (such as a C<sub>1-4</sub> haloalkoxy group such as CF<sub>3</sub>O or HCF<sub>2</sub>O), a cycloalkyl group, an amino group, an alkylthio group and a cyano group. Of these substitutable alkoxy groups, most preferred is an alkoxy group which is not substituted.

25       As the aryl moiety in the substitutable aryloxy group represented by R<sup>2</sup>, a condensation type polycyclic group such as naphthyl, as well as phenyl, may be

mentioned, and preferred is phenyl. The secondary substituent of the substitutable aryloxy group may, for example, be a halogen atom, an alkyl group, an alkoxy group or a hydroxyl group. Of these substitutable  
5 aryloxy groups, most preferred is a phenoxy group which is not substituted.

The cycloalkyl moiety in the substitutable cycloalkoxy group represented by  $R^2$  is usually one having a carbon number of from 3 to 10, and a monocyclic group  
10 such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl or cyclooctyl, as well as a condensation type polycyclic group, may, for example, be mentioned, however, preferred is a monocyclic group. The secondary substituent of the substitutable cycloalkoxy group may, for example, be a  
15 halogen, an alkyl, an alkoxy or hydroxyl. Of these substitutable cycloalkoxy groups, most preferred is a cyclohexyloxy group which is not substituted.

Here, the aryl moiety, the cycloalkyl group and the alkylthio group in the secondary substituent of the  
20 substituent represented by X,  $R^1$  and  $R^2$  are as defined for the substituents represented by X,  $R^1$ ,  $R^2$  and  $R^3$ .

The compound represented by the formula (I) may form a salt together with an acid substance, and it can form, for example, an inorganic salt such as a hydrochloride, a  
25 hydrobromate, a phosphate, a sulfate or a nitrate, or an organic salt such as an acetate, a benzoate, a p-toluenesulfonate, a methanesulfonate or a

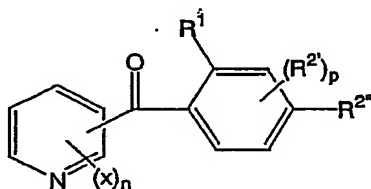
propanesulfonate.

# BEST MODE FOR CARRYING OUT THE INVENTION

Some of the preferred modes of the benzoylpyridine derivative represented by the formula (I) are shown  
 5 below. These modes may be mutually combined. Further, these compounds are useful as a fungicide.

$R^{2'}$ ,  $R^{2''}$  and  $R^{2'''}$  are as defined for the above  $R^2$ , and  $X^1$ ,  $X^2$ ,  $X^3$  and  $X^4$  are as defined for the above X.

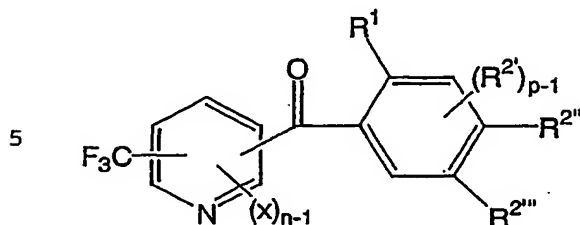
(1) A benzoylpyridine derivative represented by the  
 10 formula (I') or its salt:



15 wherein X, n and  $R^1$  are as defined in the above general formula (I),  $R^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group, p is 1, 2 or 3, and  $R^{2''}$  is a substitutable alkoxy  
 20 group or a hydroxyl group, provided that at least two of  $R^{2'}$  and  $R^{2''}$  may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a  
 25 hydroxyl group or a benzyloxy group at the 3-position; and n is 1, p is 1).

(2) The benzoylpyridine derivative or its salt

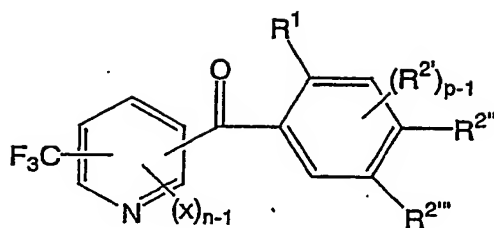
according to the above item (1), which is represented by the formula (I''):



wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4;  $\text{R}^1$  is an alkyl group;  $\text{R}^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; and each of  $\text{R}^{2''}$  and  $\text{R}^{2'''}$  is a substitutable alkoxy group.

(3) The benzoylpyridine derivative or its salt according to the above item (2), wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable cycloalkoxy group, an alkyl group, a substitutable alkylthio group or an amino group.

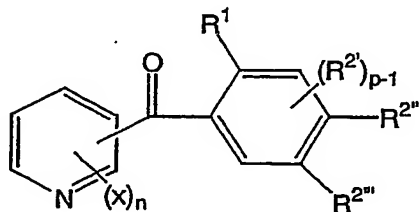
(4) The benzoylpyridine derivative or its salt according to the above item (3), which is represented by the formula (I''):



5

wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable cycloalkoxy group, an alkyl group, a substitutable alkylthio group or a substitutable amino group; n is 1, 2, 3, or 4; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; each of R<sup>2''</sup> and R<sup>2'''</sup> is a substitutable alkoxy group (excluding a case where the pyridine ring is substituted by a benzoyl group at the 3-position, and the pyridine ring has a CF<sub>3</sub> group at at least one of the 2,6-positions).

(5) The benzoylpyridine derivative or its salt according to the above item (1), which is represented by the formula (I'''):

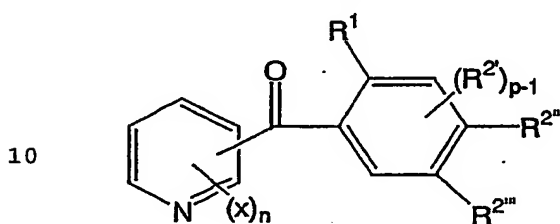


25

wherein X is a halogen atom, a substitutable alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group;

n is 1, 2, 3 or 4;  $R^1$  is an alkyl group;  $R^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is a substitutable alkoxy group.

- 5 (6) The benzoylpyridine derivative or its salt according to the above item (5), which is represented by the formula (I'''):



wherein X is a halogen atom, a substitutable alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group; n is 1, 2, 3, or 4;  $R^1$  is an alkyl group;  $R^{2'}$  is a substitutable alkyl group, a substitutable aryloxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; each of  $R^{2''}$  and  $R^{2'''}$  is a substitutable alkoxy group (excluding a case where the pyridine ring is substituted by a benzoyl group at the 3-position, and the pyridine ring has a  $CF_3$  group at at least one of the 2,6-positions).

- (7) The benzoylpyridine derivative or its salt according to the above item (5) or (6), wherein the halogen atom represented by X is a fluorine atom or a chlorine atom.

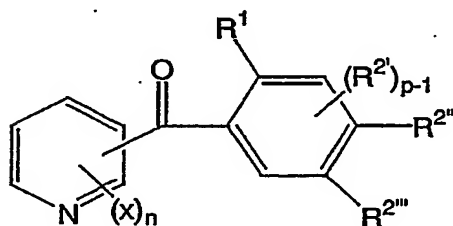
- (8) The benzoylpyridine derivative or its salt

according to the above item (5) or (6), wherein n is 3 or 4.

(9) The benzoylpyridine derivative or its salt according to the above item (5) or (6), wherein in a case where n is 1 or 2, the halogen atom represented by X is a fluorine atom or a chlorine atom.

(10) The benzoylpyridine derivative or its salt according to the above item (5), which is represented by the formula (I'''):

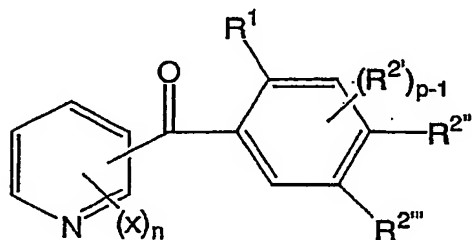
10



wherein X is a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; n is 1, 2 or 3; R¹ is an alkyl group; R²' is an alkoxy group; p is 1, 2 or 3; and each of R²'' and R²''' is an alkoxy group.

(11) The benzoylpyridine derivative or its salt according to the above item (10), which is represented by the formula (I'''):

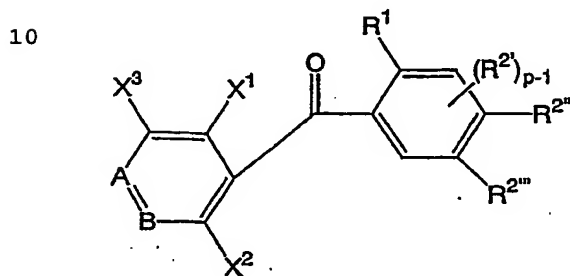
25



wherein X is a halogen atom, an alkoxy group, an alkyl

group, a  $\text{CF}_3$  group or an alkylthio group;  $n$  is 1, 2 or 3;  
 $\text{R}^1$  is an alkyl group;  $\text{R}^{2'}$  is an alkoxy group;  $p$  is 1, 2 or  
 3; and each of  $\text{R}^{2''}$  and  $\text{R}^{2'''}$  is an alkoxy group (excluding  
 a case where the pyridine ring is substituted by a  
 5 benzoyl group at the 3-position, and the pyridine ring  
 has a  $\text{CF}_3$  group at at least one of the 2,6-positions).

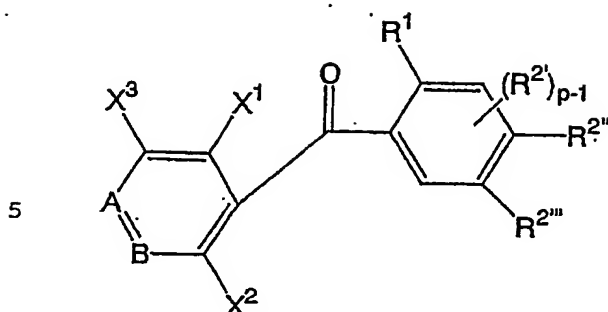
(12) The benzoylpyridine derivative or its salt  
 according to the above item (8), which is represented by  
 the formula (I'''''):



wherein B is  $-\text{CX}^4=$  when A is  $-\text{N}=$ ; B is  $-\text{N}=$  when A is  
 $-\text{CH}=$ ; each of  $\text{X}^1$  and  $\text{X}^2$  which are independent of each  
 other, is a halogen atom, an alkoxy group, an alkyl  
 group, a  $\text{CF}_3$  group or an alkylthio group;  $\text{X}^3$  is a  
 20 hydrogen atom, a halogen atom, an alkoxy group, an alkyl  
 group, a  $\text{CF}_3$  group or an alkylthio group;  $\text{X}^4$  is a  
 hydrogen atom, a halogen atom, an alkoxy group, an alkyl  
 group, a  $\text{CF}_3$  group or an alkylthio group;  $\text{R}^1$  is an alkyl  
 group;  $\text{R}^{2'}$  is an alkoxy group;  $p$  is 1, 2 or 3; and each  
 25 of  $\text{R}^{2''}$  and  $\text{R}^{2'''}$  is an alkoxy group.

(13) The benzoylpyridine derivative or its salt  
 according to the above item (8), which is represented by

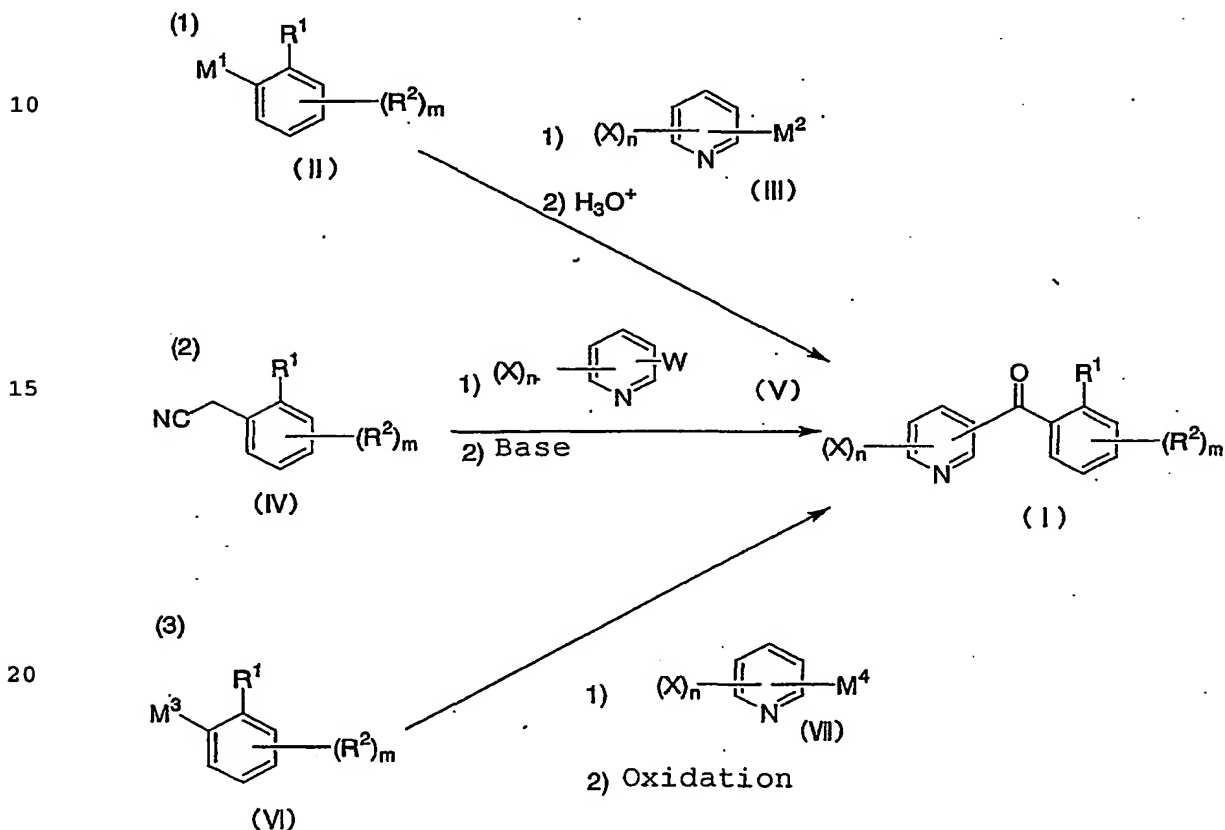
the formula (I'''''):



wherein B is -CX<sup>4</sup>= when A is -N=; B is -N= when A is -CH=; each of X<sup>1</sup> and X<sup>2</sup> which are independent of each other, is a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; X<sup>3</sup> is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; X<sup>4</sup> is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is an alkoxy group; p is 1, 2 or 3; and each of R<sup>2''</sup> and R<sup>2'''</sup> is an alkoxy group (excluding a case where A is -CH= and B is -N=, and X<sup>2</sup> is a CF<sub>3</sub> group).

The compound represented by the formula (I) or its salt may be produced in accordance with a known production process of an analogous compound (such as a process as disclosed in W096/17829). However, as the preferred modes, Processes 1 to 3 as shown in the following schemes may be mentioned. Here, X, R<sup>1</sup>, R<sup>2</sup>, n and m in the formulae are as defined above. One of the substituents represented by M<sup>1</sup> in the formula (II) and M<sup>2</sup> in the formula (III) is a cyano group, and the other is a

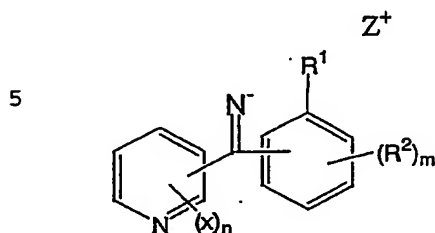
metal atom or a composite salt thereof; the substituent represented by W in the formula (V) is a halogen atom or a trifluoromethane sulfonyloxy group; one of the substituents represented by  $M^3$  in the formula (VI) and  $M^4$  in the formula (VII) is a formyl group, and the other is a metal atom or a composite salt thereof.



### Process 1

25 A process for producing the compound represented by the formula (I), which comprises subjecting a compound represented by the formula (II) and a compound presented

by the formula (III) to a condensation reaction to produce an imine compound represented by the formula (VIII):



wherein X, R<sup>1</sup>, R<sup>2</sup>, n and m are as defined above, and Z is a metal atom or a composite salt thereof, and hydrolyzing it.

10

The metal atom represented by each of M<sup>1</sup> and M<sup>2</sup> in the formulae (II) and (III) may, for example, be a typical metal atom such as lithium, magnesium, zinc or copper; or a transition metal atom such as palladium or ruthenium. Further, a composite salt of a metal atom may be used instead of a metal atom.

15

The compound of the formula (II) wherein M<sup>1</sup> is a cyano group and the compound of the formula (III) wherein M<sup>2</sup> is a cyano group may be produced in accordance with a known process such as a process as disclosed in Journal of the Chemical Society, Perkin transactions 1 pages 2323-2326, 1999.

20

The condensation reaction to produce an imine compound is carried out in the presence of a proper solvent (such as an inert solvent such as tetrahydrofuran, diethyl ether, dimethoxyethane, hexane, benzene, toluene or methylene chloride, or a mixed

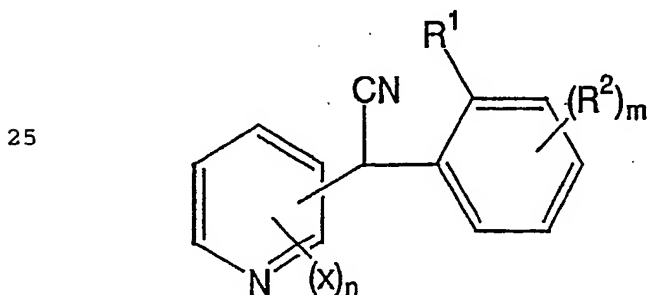
25

solvent thereof) at a reaction temperature of from -100 to 70°C, preferably from -80 to 30°C. This reaction is carried out preferably in an inert gas atmosphere of e.g. nitrogen or argon.

5       The imine compound produced by the condensation reaction is hydrolyzed by a known procedure and converted into the compound represented by the formula (I). The hydrolysis reaction may be carried out in the presence of e.g. water, an alcohol or a mixture thereof. Here, in  
10   Process 1, the condensation reaction and the hydrolysis reaction are usually carried out continuously, and no imine compound is isolated. Further, to obtain the compound represented by the formula (I) with a high yield, it is preferred to carry out the hydrolysis  
15   reaction after the condensation reaction is completely conducted.

#### Process 2

A process for producing the compound represented by the formula (I), which comprises subjecting a compound  
20   represented by the formula (IV) and a compound represented by the formula (V) to a condensation reaction to produce a compound represented by the formula (IX):



wherein X, R<sup>1</sup>, R<sup>2</sup>, n and m are as defined above, and  
subjecting it to decyanogenation oxidatively in the  
presence of a base.

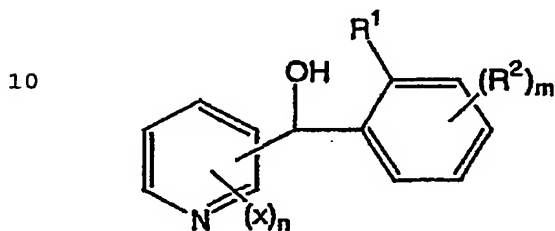
The reaction to produce the compound represented by  
5 the formula (IX) at the first half stage of Process 2 is  
carried out usually in the presence of a base preferably  
in a solvent. The base to be used for the reaction may,  
for example, be lithium hydride, sodium hydride, sodium  
methoxide, sodium ethoxide or potassium tert-butoxide.  
10 The solvent may, for example, be tetrahydrofuran, diethyl  
ether, benzene, toluene, methylene chloride, chloroform  
or DMF, or a mixed solvent thereof. This reaction is  
carried out preferably at a reaction temperature of from  
0 to 100°C. Further, it is carried out preferably in an  
15 inert gas atmosphere of e.g. nitrogen or argon. Further,  
sodium benzenesulfinate or sodium p-toluenesulfinate may  
be added as the case requires to accelerate the reaction.

The oxidative decyanogenation reaction at the last  
half stage in Process 2 is carried out in the presence of  
20 a base. The base may, for example, be sodium hydride,  
potassium hydride, sodium carbonate or potassium  
carbonate. Further, a phase-transfer catalyst (such as  
benzyl triethylammonium chloride or tetrabutylammonium  
hydrogensulfate) may be used as the case requires. This  
25 reaction is usually carried out in a proper solvent (such  
as an inert solvent such as methylene chloride,  
chloroform, 1,2-dichloroethane, benzene, toluene, DMF or

DMSO, or a water-containing solvent or a mixed solvent thereof) at a reaction temperature of from 0 to 50°C.

### Process 3

A process for producing the compound represented by the formula (I), which comprises reacting a compound represented by the formula (VI) and a compound represented by the formula (VII) to produce phenylpyridyl methanol represented by the formula (X):



15 (wherein X, n, m, R¹ and R² are as defined above with a proviso as in formula (I)) and oxidizing it.

Each of metal atoms represented by M³ and M⁴ in Process 3, may, for example, be a typical metal atom such as lithium, magnesium, zinc or copper; or a transition metal atom such as palladium or ruthenium. Further, a composite salt of a metal atom may be used instead of a metal atom.

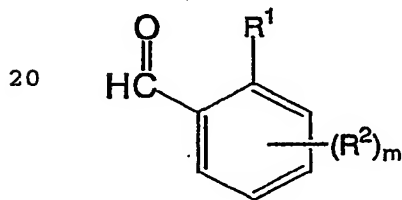
The compound of the formula (VI) wherein the substituent represented by M³ is a formyl group and the compound of the formula (VII) wherein the substituent represented by M⁴ is a formyl group, may usually be produced in accordance with a known process such as a

process as disclosed in Journal of Organic Chemistry vol. 57, pages 6847-6852, 1992.

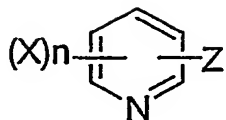
The phenylpyridyl methanol represented by the formula (X), formed from the compound represented by the formula (VI) and the compound represented by the formula (VII), may be oxidized by a known means such as a metal oxidizing agent such as manganese dioxide or chromic acid, a Swern oxidation method (dimethylsulfoxide + oxalyl chloride) or a ruthenium oxidation method (tetrapropylammonium perruthenate + N-methylmorpholine-N-oxide) and converted to a compound represented by the formula (I)

Now, mode of carrying out Process 3 is described below.

(1) A process for producing the compound represented by the formula (I), which comprises reacting a substituted benzaldehyde represented by the formula (VI-1):

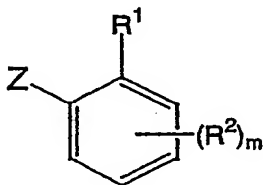


(wherein  $R^1$ ,  $R^2$  and  $m$  are as defined above), and a metal salt of a substituted pyridine derivative represented by the formula (VII-1):



(wherein X is as defined above, and Z is a metal atom or  
 5 a composite salt thereof), to produce phenylpyridyl  
 methanol represented by the formula (X), and oxidizing  
 it.

(2) A process for producing the compound represented  
 by the formula (I), which comprises reacting a metal salt  
 10 of a substituted benzene derivative represented by the  
 formula (VI-2):



15

(wherein  $R^1$ ,  $R^2$  and  $m$  are as defined above, and Z is a  
 metal atom or a composite salt thereof), and a  
 substituted pyridylaldehyde represented by the formula  
 (VII-2):

20

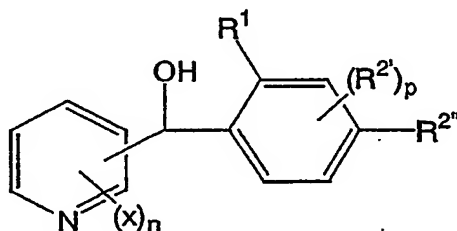


(wherein X is as defined above), to produce phenylpyridyl  
 25 methanol represented by the formula (X), and oxidizing  
 it.

Here, the preferred modes of the phenylpyridyl

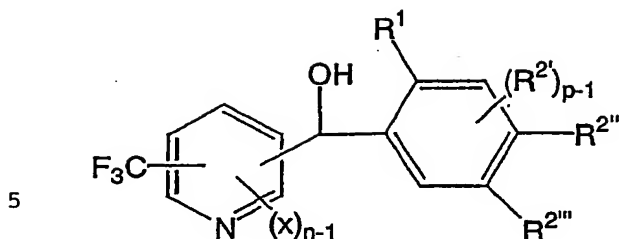
methanol represented by the formula (IX) which is an intermediate for production of the compound represented by the formula (I) are shown below.

(1) Phenylpyridyl methanol represented by the  
5 formula (X'):



10 wherein X, n and R<sup>1</sup> are as defined for the above general formula (I), R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group, p is 1, 2 or 3; and R<sup>2''</sup> is a substitutable alkoxy  
15 group or a hydroxyl group, provided that at least two of R<sup>2'</sup> and R<sup>2''</sup> may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a  
20 hydroxyl group or a benzyloxy group at the 3-position; and n is 1, p is 1).

(2) The phenylpyridyl methanol according to the above item (1), which is represented by the formula (X''):



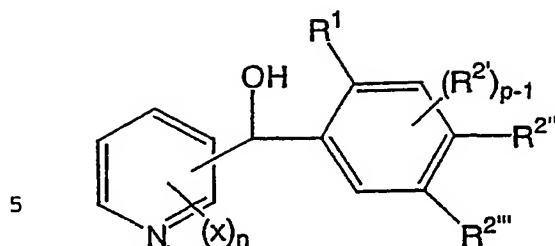
wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group or a substitutable cycloalkoxy group, p is 1, 2 or 3, and each of R<sup>2''</sup> and R<sup>2'''</sup> is a substitutable alkoxy group.

(3) The phenylpyridyl methanol according to the above item (2), wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable cycloalkoxy group, an alkyl group, a substitutable alkylthio group or a substitutable amino group.

(4) The phenylpyridyl methanol according to the above item (2) or (3), wherein the pyridine ring is substituted by a benzoyl group at the 4-position.

(5) The phenylpyridyl methanol according to the above item (1), which is represented by the formula

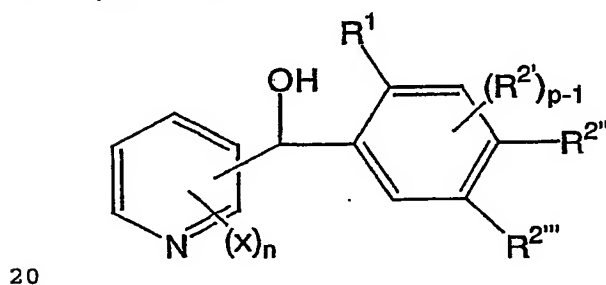
(X''):



wherein X is a halogen atom, a substitutable alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; n is 1, 2, 3 or 4; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; and each of R<sup>2''</sup> and R<sup>2'''</sup> is a substitutable alkoxy group.

(6) The phenylpyridyl methanol according to the above item (5), which is represented by the formula

15 (X'''):



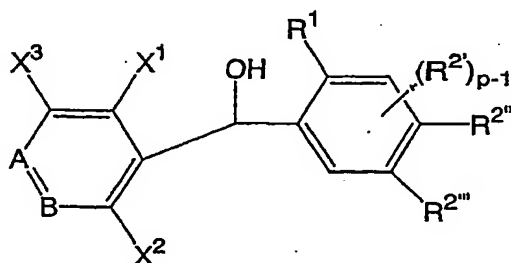
wherein X is a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; n is 1, 2 or 3; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is an alkoxy group; p is 1, 2 or 3; and each of R<sup>2''</sup> and R<sup>2'''</sup> is an alkoxy group.

(7) The phenylpyridyl methanol according to the above item (5) or (6), wherein the pyridine ring is

substituted by a benzoyl group at the 4-position.

(8) The phenylpyridyl methanol according to the above item (6), which is represented by the formula (X'''''):

5



10

wherein B is  $-CX^4=$  when A is  $-N=$ ; B is  $-N=$  when A is  $-CH=$ ; each of  $X^1$  and  $X^2$  which are independent of each other, is a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $X^3$  is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $X^4$  is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $R^1$  is an alkyl group;  $R^{2'}$  is an alkoxy group; p is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is an alkoxy group.

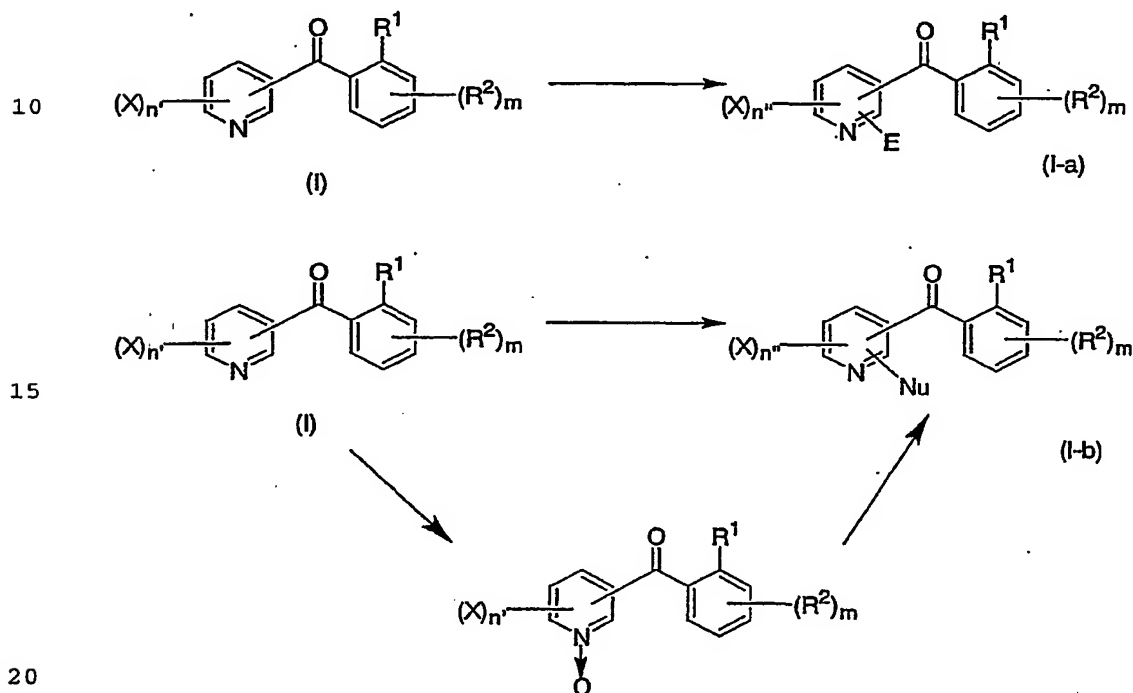
(9) The phenylpyridyl methanol according to the above item (8), wherein A is  $-N=$ .

A substituent may further be introduced into the compound represented by the formula (I) electrophilically or nucleophilically. That is, the compound represented by the formula (I) may be converted into a compound

25

represented by the formula (I-a) or (I-b) as illustrated in the following scheme. Further, it is also possible to radically introduce a substituent into the compound represented by the formula (I). Here, in the formula (I-a), E is an electrophilic reagent, and in the formula (I-b), Nu is a nucleophilic reagent.

$n'$  and  $n''$  are as defined for the above  $n$ .

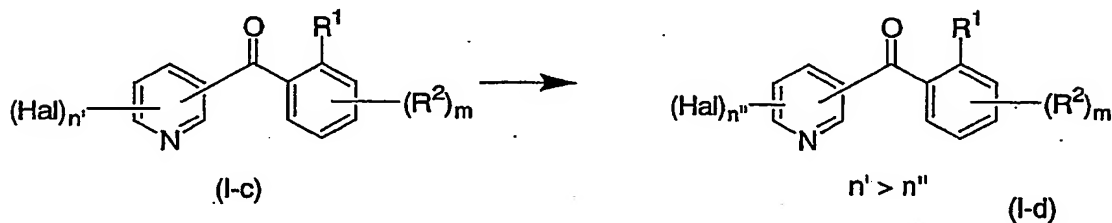


$$n' \geq n''$$

The reaction to prepare the compound represented by the formula (I-a) varies properly depending upon the electrophile, and the reaction may usually be carried out by a known process or a process in accordance therewith. For example, the above-described Process 1 may be

employed. The nucleophilic substitution to prepare the compound represented by the formula (I-b) varies properly depending upon the nucleophile, and the reaction may usually be carried out by a known process or a process in accordance therewith. For example, in a case of an ethyloxy nucleophilic reagent, it is preferred to carry out the reaction in the presence of an inert solvent such as ethanol or dioxane, toluene or octane as the solvent, at a reaction temperature of from 0 to 120°C for a proper time. The ethyloxy nucleophilic reagent is used in from 0.1 to 10 mol equivalent amount, preferably in from 0.5 to 5 mol equivalent amount.

Further, the compound represented by the formula (I-c) (the compound of the formula (I) wherein X is a halogen atom) may further be converted into a compound represented by the formula (I-d) by removing the halogen substituent, as shown in the following scheme. For the reaction as illustrated by the scheme, catalytic hydrogenation, hydrogen transfer reaction or metal reduction reaction may properly be employed. In the scheme, Hal is a halogen atom.



The catalytic hydrogenation may be carried out in

the presence of a catalyst under hydrogen gas atmosphere under normal pressure or under elevated pressure in the presence of a proper solvent. The catalyst to be used may, for example, be a catalyst system having platinum, 5 palladium, rhodium, ruthenium, nickel or iridium. The solvent to be used may, for example, be water, an alcohol (such as methanol or ethanol), ethyl acetate, acetic acid, dioxane, ether, benzene or hexane. In such a case, the catalyst is used in a proportion of from 0.01 to 1.2 10 mol based on the compound represented by the formula (I-c). Further, the reaction may be carried out in the presence of a base such as triethylamine or sodium hydrogen carbonate. Further, a known reduction reaction may be employed such as hydrogen transfer reaction (e.g. 15 palladium carbon, ammonium formate as a hydrogen source, or sodium dihydrogen phosphate) or a metal reduction reaction (e.g. samarium diiodide).

Now, specific Synthesis Examples of the benzoylpyridine derivative represented by the formula (I) 20 and the intermediate for its production are described below (the compounds in Synthesis Examples are based on IUPAC nomenclature, and the substitution positions may be different from those shown in Tables as mentioned hereinafter, expediently).

#### 25 SYNTHESIS EXAMPLE 1

Synthesis of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2,6-dichloro-4-trifluoromethylpyridine (compound No. 3)

(a) 14 ml (20 mmol) of n-butyllithium (1.5 M hexane solution) was dropwise added at 0°C to a solution having 2.9 ml (21 mmol) of diisopropylamine dissolved in 62 ml of tetrahydrofuran, followed by stirring for 30 minutes.

5 The solution was cooled to -20°C, a solution having 4.0 g (19 mmol) of 2,6-dichloro-4-trifluoromethylpyridine dissolved in 5 ml of tetrahydrofuran was added thereto, followed by stirring for 5 minutes, and a solution having 3.8 g (18 mmol) of 2,3,4-trimethoxy-6-methylbenzaldehyde

10 dissolved in 7 ml of tetrahydrofuran was added thereto, followed by stirring for 1.5 hours. 30 ml of water was added to the mixture to terminate the reaction, and tetrahydrofuran was distilled off under reduced pressure. Extraction with ethyl acetate was carried out, the

15 organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 6.2 g (yield 81%) of (2,3,4-trimethoxy-6-

20 methylphenyl)(2,6-dichloro-4-trifluoromethyl-3-pyridyl)methanol (brown oily substance).

(b) 14 g of manganese dioxide was added to a solution having 5.4 g of (2,3,4-trimethoxy-6-methylphenyl)(2,6-dichloro-4-trifluoromethyl-3-

25 pyridyl)methanol obtained in step (a) dissolved in 140 ml of toluene, followed by stirring under reflux by heating for 6 hours. The mixture was cooled and then subjected

to filtration, and toluene was distilled off under reduced pressure to obtain 4.4 g (yield 81%) of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2,6-dichloro-4-trifluoromethylpyridine (compound No. 3; m.p. 81-83°C).

5 SYNTHESIS EXAMPLE 2

Synthesis of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine (compound No. 11) and 3-(2,3,4-trimethoxy-6-methylbenzoyl)-4-trifluoromethylpyridine (compound No. 7)

10 2.4 ml (17 mmol) of triethylamine and 0.3 g of 5% palladium carbon were added to a solution having 3.4 g (8.0 mmol) of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2,6-dichloro-4-trifluoromethylpyridine (compound No. 3) obtained in Synthesis Example 1 dissolved in 50 ml of  
15 methanol, followed by stirring under hydrogen atmosphere for 6.5 hours. The mixture was subjected to filtration, 50 ml of water was added thereto, and methanol was distilled off under reduced pressure. Extraction of ethyl acetate was carried out, the organic layer was  
20 dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain  
25 1.7 g (yield 55%) of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine (compound No. 11; m.p. 110-112°C) and 1.1 g (yield 37%) of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-4-

trifluoromethylpyridine (compound No. 7; m.p. 59-62°C).

### SYNTHESIS EXAMPLE 3

#### Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,5-dichloro-3-trifluoromethylpyridine (compound No. 90)

5 (a) 17 ml (25 mmol) of n-butyllithium (1.5 M hexane solution) was dropwise added at 0°C to a solution having 3.6 ml (25 mmol) of diisopropylamine dissolved in 60 ml of diethyl ether, followed by stirring for 45 minutes. The solution was cooled to -78°C, a solution having 6.0 g  
10 (24 mmol) of 2,3,6-trichloro-5-trifluoromethylpyridine dissolved in 8 ml of diethyl ether was added thereto, followed by stirring for 5 minutes, and a solution having 5.0 g (24 mmol) of 2,3,4-trimethoxy-6-methylbenzaldehyde dissolved in 12 ml of toluene was added thereto, followed  
15 by stirring for 1 hours. 30 ml of water was added to the mixture to terminate the reaction, the aqueous layer was extracted with ethyl acetate, and then the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under  
20 reduced pressure to obtain (2,3,4-trimethoxy-6-methylphenyl)(2,3,6-trichloro-5-trifluoromethyl-4-pyridyl)methanol (m.p. 131-135°C).

(b) 2.7 ml (19 mmol) of triethylamine and 0.9 g of 5% palladium carbon were added to a solution having (2,3,4-trimethoxy-6-methylphenyl)(2,3,6-trichloro-5-trifluoromethyl-4-pyridyl)methanol obtained in step (a)  
25 dissolved in 200 ml of methanol, followed by stirring

under hydrogen atmosphere for 14 hours. The mixture was subjected to filtration, 30 ml of water was added thereto, and methanol was distilled off under reduced pressure. Extraction with ethyl acetate was carried out, the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 2.38 g (yield 24%) of (2,3,4-trimethoxy-6-methylphenyl)(2,5-dichloro-3-trifluoromethyl-4-pyridyl)methanol (m.p. 162-165°C).

(c) 14 g of manganese dioxide was added to a solution having 3.5 g (8.2 mmol) of (2,3,4-trimethoxy-6-methylphenyl)(2,5-dichloro-3-trifluoromethyl-4-pyridyl)methanol obtained in step (b) dissolved in 100 ml of toluene, followed by stirring under reflux by heating for 6 hours. The mixture was cooled and then subjected to filtration, and toluene was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 3.1 g (yield 89%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,5-dichloro-3-trifluoromethylpyridine (compound No. 90; m.p. 106-109°C).

#### SYNTHESIS EXAMPLE 4

Synthesis of 3-(4,5-dimethoxy-2-methylbenzoyl)-2-methoxy-4-trifluoromethylpyridine (compound No. 32)

0.9 g (16 mmol) of sodium methoxide was added to a

solution having 1.5 g (4.2 mmol) of 3-(4,5-dimethoxy-2-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine synthesized in accordance with a process in Synthesis Example 1 dissolved in 20 ml of toluene, followed by stirring under reflux by heating for 4 hours. The mixture was cooled, and then 20 ml of water was added thereto to terminate the reaction, the aqueous solution was extracted with ethyl acetate, and the organic layer was dried over anhydrous sodium sulfate and subjected to filtration by using a silica gel cake. The solvent was distilled off under reduced pressure to obtain 1.5 g (yield 99%) of 3-(4,5-dimethoxy-2-methylbenzoyl)-2-methoxy-4-trifluoromethylpyridine (compound No. 32; m.p. 125-127°C).

#### SYNTHESIS EXAMPLE 5

##### Synthesis of 3-[4,5-(methylenedioxy)-2-methylbenzoyl]-2-chloro-4-trifluoromethylpyridine (compound No. 13)

(a) 3.2 ml (62 mmol) of bromine was dropwise added at 0°C to a solution having 7.0 ml (58 mmol) of 3,4-(methylenedioxy)toluene and 5.5 ml (68 mmol) of pyridine dissolved in 110 ml of dichloromethane, followed by stirring for 30 minutes, and the temperature was raised to room temperature, followed by stirring for 22 hours. The mixture was washed with an aqueous sodium hydroxide solution, dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus

obtained was purified by silica gel column chromatography to obtain 13 g (yield 99%) of 2-bromo-4,5-(methylenedioxy)toluene.

(b) 13 mL (20 mmol) of n-butyllithium (1.5 M hexane solution) was dropwise added at -78°C to a solution having 4.0 g (19 mmol) of 2-bromo-4,5-(methylenedioxy)toluene dissolved in 50 mL of tetrahydrofuran, followed by stirring for 30 minutes, and 1.5 mL (19 mmol) of dimethylformamide was added thereto, followed by stirring for 70 minutes. 30 mL of water was added to the mixture to terminate the reaction, and tetrahydrofuran was distilled off under reduced pressure. Extraction with chloroform was carried out, the organic layer was dried over anhydrous sodium sulfate and subjected to filtration by using a silica gel cake, and the solvent was distilled off under reduced pressure to obtain 3.1 g (yield 99%) of 2-methyl-4,5-(methylenedioxy)benzaldehyde (m.p. 84-86°C).

(c) Using 1.5 g (8.3 mmol) of 2-chloro-4-trifluoromethylpyridine and 1.4 g (8.2 mmol) of 2-methyl-4,5-(methylenedioxy)benzaldehyde, 2.1 g (yield 73%) of (2-methyl-4,5-(methylenedioxy)phenyl)(2-chloro-4-trifluoromethyl-3-pyridyl)methanol (m.p. 127-130°C) was obtained by a process in accordance with step (a) of Synthesis Example 1.

(d) Using 1.5 g (4.3 mmol) of (2-methyl-4,5-(methylenedioxy)phenyl)(2-chloro-4-trifluoromethyl-3-

pyridyl)methanol obtained in step (c) and 8.0 g (92 mmol) of manganese dioxide, 0.3 g (yield 22%) of 3-[4,5-(methylenedioxy)-2-methylbenzoyl]-2-chloro-4-trifluoromethylpyridine (compound No. 13; m.p. 119-122°C) was obtained by a process in accordance with step (b) of Synthesis Example 1.

## SYNTHESIS EXAMPLE 6

Synthesis of 3-(5-benzyloxy-4-methoxy-2-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine (compound No. 27)

10 (a) A dimethylformamide (15 mL) solution of 2-methoxy-4-methylphenol (6.91 g) was dropwise added to a dimethylformamide (20 mL) suspension of sodium hydride (2.4 g) under cooling with ice, followed by stirring for 30 minutes. A dimethylformamide (15 mL) solution of  
15 benzyl bromide (9.41 g) was dropwise added thereto, and tetrabutylammonium bromide in a catalytic amount was added thereto, followed by stirring at the same temperature for 30 minutes. The temperature was raised to room temperature and stirring was further carried out  
20 for one night. The reaction solution was poured into water (250 mL), and extraction with ethyl acetate (100 mL) was carried out three times. The ethyl acetate phase was washed with water (100 mL) three times and then washed with an aqueous sodium chloride solution (100 mL).  
25 After drying over magnesium sulfate, the solvent was distilled off under reduced pressure, the residue was purified by silica gel column chromatography (hexane-

ethyl acetate) to obtain 11.4 g of 4-benzyloxy-3-methoxytoluene (m.p. 38-39°C) quantitatively, and its structure was confirmed by nuclear magnetic resonance spectrum.

5 (b) 4-benzyloxy-3-methoxytoluene (8.0 g) was dissolved in dimethylformamide (30 ml), and a dimethylformamide (15 ml) solution of N-bromosuccinimide (6.36 g) was dropwise added thereto, followed by stirring at room temperature for one night. The reaction solution  
10 was poured into ice water (400 ml), and crystals thus deposited were collected by filtration, adequately washed with water, and dried for one night to obtain 10.64 g of 4-benzyloxy-2-bromo-5-methoxytoluene (m.p. 110-111°C) substantially quantitatively, and its structure was  
15 confirmed by nuclear magnetic resonance spectrum.

(c) A hexane solution (17 ml) of n-butyllithium was dropwise added to a tetrahydrofuran (190 ml) solution of 4-benzyloxy-2-bromo-5-methoxytoluene (7.83 g) at -78°C over a period of 20 minutes, followed by stirring at the  
20 same temperature for 1 hour. A tetrahydrofuran (10 ml) solution of dimethylformamide (3.73 g) was dropwise added thereto at -78°C, followed by stirring at the same temperature for 1 hour. The temperature was gradually raised to room temperature, and stirring was further  
25 carried out for one night. The reaction solution was poured into an aqueous ammonium chloride solution (200 ml), and extraction with ethyl acetate (150 ml) was

carried out twice. The ethyl acetate phase was washed with an aqueous sodium chloride solution (100 ml) twice and dried over magnesium sulfate, and the solvent was distilled off under reduced pressure. The residue was  
5 purified by silica gel column chromatography (hexane-ethyl acetate) to obtain 3.14 g (yield 48%) of 5-benzyloxy-4-methoxy-2-methylbenzaldehyde (m.p. 107-109°C), and its structure was confirmed by nuclear magnetic resonance spectrum.

10 (d) A hexane solution (11.4 ml) of n-butyllithium was dropwise added to a tetrahydrofuran (45 ml) solution of diisopropylamine (2.81 g) at 0°C, followed by stirring for 1 hour to prepare a tetrahydrofuran solution of lithium diisopropylamide. The solution was cooled to  
15 -50°C, and a tetrahydrofuran (7.5 ml) solution of 2-chloro-4-trifluoromethylpyridine (2.81 g) was gradually added thereto, followed by stirring at the same temperature for 30 minutes. The solution was cooled to -78°C, and a tetrahydrofuran (37.5 ml) solution of 5-  
20 benzyloxy-4-methoxy-2-methylbenzaldehyde (3.97 g) was gradually added thereto, followed by stirring at the same temperature for 1 hour. A saturated aqueous ammonium chloride solution (50 ml) was added thereto, the temperature was raised to room temperature, the mixture  
25 was poured into a saturated aqueous sodium bicarbonate solution (50 ml), and extraction with ethyl acetate (150 ml) was carried out twice. The ethyl acetate phase was

washed with an aqueous sodium chloride solution (100 ml) and dried over magnesium sulfate, and the solvent was distilled off under reduced pressure. The residue was purified by silica gel column chromatography (hexane-ethyl acetate) to obtain 6.48 g (yield 96%) of (5-benzyloxy-4-methoxy-2-methylphenyl) (2-chloro-4-trifluoromethyl-3-pyridyl)methanol as a red-yellow oily substance, and its structure was confirmed by nuclear magnetic resonance spectrum.

10 (e) (5-benzyloxy-4-methoxy-2-methylphenyl) (2-chloro-4-trifluoromethyl-3-pyridyl)methanol (5.9 g) was dissolved in a mixed solvent of anhydrous methylene chloride (50 ml) and acetonitrile (5 ml), and tetrapropylammonium perruthenate (95 mg), N-methylmorpholine-N-oxide (2.38 g) and molecular sieve 4A  
15 (6.8 g) were sequentially added thereto, followed by stirring in a stream of argon at room temperature for three nights. The reaction mixture was distilled off under reduced pressure, the residue thus obtained was  
20 suspended in methylene chloride and subjected to filtration by celite, and the residue was adequately washed with methylene chloride (200 ml). The solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (hexane-ethyl acetate) to obtain 4.93 g (yield 84%) of 3-(5-benzyloxy-4-methoxy-2-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine (compound No. 27; m.p. 116-

117°C), and its structure was confirmed by nuclear magnetic resonance spectrum.

#### SYNTHESIS EXAMPLE 7

Synthesis of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2-methylthio-4-trifluoromethylpyridine (compound No. 50)

Sodium methanethiolate (0.32 g) was added to a dimethylformamide (15 ml) solution of 0.9 g of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2-chloro-4-trifluoromethylpyridine (compound No. 11) at room temperature, followed by stirring for 1 hour. The mixture was poured into water (50 ml), and extraction with ethyl acetate was carried out. The ethyl acetate phase was dried over sodium sulfate, the solvent was distilled off under reduced pressure, and the residue was purified by silica gel column chromatography (hexane-ethyl acetate) to obtain 0.54 g (yield 58%) of 3-(2,3,4-trimethoxy-6-methylbenzoyl)-2-methylthio-4-trifluoromethylpyridine (compound No. 50; pale yellow oily substance), and its structure was confirmed by nuclear magnetic resonance spectrum.

#### SYNTHESIS EXAMPLE 8

Synthesis of 5-(2,3,4-trimethoxy-6-methylbenzoyl)-3-acetyl-2,6-dichloro-4-trifluoromethylpyridine (compound No. 62)

(a) 9.6 ml (14 mmol) of n-butyllithium (1.5 M hexane solution) was dropwise added to a tetrahydrofuran (16 ml) solution of 2.0 ml (14 mmol) of diisopropylamine at 0°C,

followed by stirring for 30 minutes. The solution was cooled to  $-50^{\circ}\text{C}$ , a tetrahydrofuran (11 mL) solution of 2.9 g (7 mmol) of (2,3,4-trimethoxy-6-methylphenyl)(2,6-dichloro-4-trifluoromethyl-3-pyridyl)methanol was added thereto, followed by stirring for 30 minutes, then the solution was cooled to  $-78^{\circ}\text{C}$ , and acetaldehyde in an excess amount was added thereto, followed by stirring for 2 hours. 30 mL of water was added to the mixture to terminate the reaction, and tetrahydrofuran was distilled off under reduced pressure. Extraction with ethyl acetate was carried out, the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 2.5 g (yield 78%) of (2,3,4-trimethoxy-6-methylphenyl)(2,6-dichloro-5-(1-hydroxyethyl)-4-trifluoromethyl-3-pyridyl)methanol.

(b) 10 g of manganese dioxide was added to a toluene (80 mL) solution of 2.3 g (5 mmol) of (2,3,4-trimethoxy-6-methylphenyl)(2,6-dichloro-5-(1-hydroxyethyl)-4-trifluoromethyl-3-pyridyl)methanol obtained in step (a), followed by stirring under reflux by heating for 1 hour. The reaction solution was cooled to room temperature and then subjected to filtration, and toluene was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 1.5 g (yield 66%) of 5-(2,3,4-trimethoxy-6-

methylbenzoyl)-3-acetyl-2,6-dichloro-4-trifluoromethylpyridine (compound No. 62; m.p. 109-112°C).

SYNTHESIS EXAMPLE 9

5 Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2-chloro-3-trifluoromethyl-5-methoxypyridine (compound No. 123)

(a) 70.0 ml (106 mmol) of n-butyllithium (1.5 M hexane solution) was dropwise added to a diethyl ether.  
10 120 ml solution of 15.0 ml (107 mmol) of diisopropylamine at 0°C, followed by stirring for 1 hour. The solution was cooled to -78°C, a diethyl ether 10 ml solution of 22.1 g (102 mmol) of 2,3-dichloro-5-trifluoromethylpyridine was added thereto, followed by  
15 stirring for 30 minutes, and then a toluene 40 ml solution of 21.0 g (100 mmol) of 2,3,4-trimethoxy-6-methylbenzaldehyde was added thereto, followed by stirring for 2 hours. 30 ml of water was added to the mixture to terminate the reaction, the aqueous layer was  
20 extracted with ethyl acetate, and then the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain  
25 24.8 g (yield 58%) of (2,3,4-trimethoxy-6-methylphenyl) (2,3-dichloro-5-trifluoromethyl-4-pyridyl)methanol (m.p. 95-98°C).

(b) 2.1 g of 5% palladium carbon was added to a methanol 200 ml solution of 24.8 g (58.1 mmol) of (2,3,4-trimethoxy-6-methylphenyl) (2,3-dichloro-5-trifluoromethyl-4-pyridyl)methanol obtained in step (a) and 9.50 ml (68.2 mmol) of triethylamine, followed by stirring under hydrogen atmosphere for 4 hours. The mixture was subjected to filtration, 50 ml of water was added thereto, and methanol was distilled off under reduced pressure. The aqueous layer was extracted with ethyl acetate, and the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 15.9 g (yield 70%) of (2,3,4-trimethoxy-6-methylphenyl) (3-chloro-5-trifluoromethyl-4-pyridyl)methanol (m.p. 102-105°C).

(c) 45 g of manganese dioxide was added to a toluene 220 ml solution of 15.9 g (40.6 mmol) of (2,3,4-trimethoxy-6-methylphenyl) (3-chloro-5-trifluoromethyl-4-pyridyl)methanol obtained in step (b), followed by stirring under reflux by heating for 2 hours. The mixture was subjected to filtration, and the solvent was distilled off under reduced pressure to obtain 14.9 g (yield 94%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-trifluoromethylpyridine (compound No. 102; m.p. 75-77°C).

(d) 16.4 g (304 mmol) of sodium methoxide was added

to a toluene 150 ml solution of 18.5 g (47.5 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-trifluoromethylpyridine obtained in step (c) and 16.6 ml (95.4 mmol) of hexamethylphosphoric triamide, followed by  
5 stirring under reflux by heating for 30 minutes. Water was added thereto to terminate the reaction, the aqueous layer was extracted with ethyl acetate, and the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled  
10 off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 11.7 g (yield 64%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-methoxy-5-trifluoromethylpyridine (compound No. 122; m.p. 103-106°C).

15 (e) 6.1 g (28 mmol) of m-chloroperbenzoic acid (m-CPBA) was added to a chloroform 100 ml solution of 5.6 g (15 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-methoxy-5-trifluoromethylpyridine (compound No. 122) at 0°C, followed by stirring at room temperature for 18  
20 hours. The reaction solution was washed with an aqueous sodium hydroxide solution, and the solvent was distilled off under reduced pressure to obtain 5.8 g (yield 99%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-methoxy-5-trifluoromethylpyridine-N-oxide (m.p. 128-134°C).

25 (f) 1.8 ml (19 mmol) of phosphorus oxychloride was added to 4 ml of toluene and 8 ml of dimethylformamide at 0°C, followed by stirring for 10 minutes, and 4.0 g (10

mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-methoxy-5-trifluoromethylpyridine-N-oxide was added thereto, followed by stirring for 20 minutes. Stirring was carried out at room temperature for 2 hours, and then the reaction solution was poured into ice water to terminate the reaction. The aqueous layer was extracted with ethyl acetate, and then the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure.

10 The crude product thus obtained was purified by silica gel column chromatography to obtain 3.57 g (yield 85%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2-chloro-3-trifluoromethyl-5-methoxypyridine (compound No. 123; m.p. 117-119°C).

15 SYNTHESIS EXAMPLE 10

Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2-bromo-3-trifluoromethyl-5-methoxypyridine (compound No. 124)

Using 7.2 g (18 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-methoxy-5-trifluoromethylpyridine-N-oxide, 7 ml of toluene, 17 ml of dimethylformamide and 10 g (35 mmol) of phosphorus oxybromide, 4.1 g (yield 49%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2-bromo-3-trifluoromethyl-5-methoxypyridine (compound No. 124; m.p. 145-147°C) was obtained in the same process as in Synthesis Example 9 step (f).

20

25

## SYNTHESIS EXAMPLE 11

Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,3,5-trichloropyridine (compound No. 186)

(a) 17.2 ml (26.7 mmol) of n-butyllithium (1.56 M  
5 hexane solution) was dropwise added to a diethyl ether  
(20 ml) solution of 2.7 g (26.7 mmol) of diisopropylamine  
at 0°C, followed by stirring for 1 hour. The solution  
was cooled to -78°C, a toluene solution of 4.8 g (26.7  
mmol) of 2,3,5-trichloropyridine was dropwise added  
10 thereto, and then a toluene solution of 5.0 g (24.0 mmol)  
of 2,3,4-trimethoxy-6-methylbenzaldehyde was dropwise  
added thereto, followed by stirring for 30 minutes. The  
temperature was recovered to room temperature, and  
stirring was carried out further for 1 hour. 30 ml of  
15 water was added to the mixture to terminate the reaction,  
and ethyl acetate was added for extraction. The organic  
layer was dried over anhydrous sodium sulfate and  
subjected to filtration, and the solvent was distilled  
off under reduced pressure. The crude product thus  
20 obtained was purified by silica gel column chromatography  
to obtain 6.7 g (yield 72%) of amorphous (2,3,4-  
trimethoxy-6-methylphenyl)(2,3,5-trichloro-4-  
pyridyl)methanol.

(b) 16.2 g of manganese dioxide was added to a  
25 toluene (180 ml) solution of 5.6 g of (2,3,4-trimethoxy-  
6-methylphenyl)(2,3,5-trichloro-4-pyridyl)methanol  
obtained in step (a), followed by stirring under reflux

by heating for 3 hours. After the mixture was cooled, it was subjected to filtration, and the solvent was distilled off under reduced pressure to obtain 4.7 g (yield 87%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,3,5-trichloropyridine (compound No. 186; m.p. 60-61°C).

## SYNTHESIS EXAMPLE 12

Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3,5-dichloropyridine (compound No. 191)

4.6 g (6.9 mmol) of triethylamine and 1.8 g of 10% palladium carbon were added to a methanol 280 ml solution of 17.8 g (4.6 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,3,5-trichloropyridine (compound No. 186), followed by stirring under hydrogen atmosphere at room temperature for 7 hours. The mixture was subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 11.6 g (yield 72%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3,5-dichloropyridine (compound No. 191; m.p. 109-111°C).

## SYNTHESIS EXAMPLE 13

Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-methoxypyridine (compound No. 244)

5.0 g (2.8 mmol) of hexamethylphosphoric triamide and 1.1 g (2.1 mmol) of sodium methoxide were added to a toluene (60 ml) solution of 5.0 g (1.4 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3,5-dichloropyridine

(compound No. 191), followed by stirring under reflux by heating for 5 hours. After the mixture was cooled, 50 ml of water was added to the mixture to terminate the reaction, and ethyl acetate was added thereto for  
5 extraction. The organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 3.4 g (yield 69%) of 4-  
10 (2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-methoxypyridine (compound No. 244; pale yellow oily substance).

#### SYNTHESIS EXAMPLE 14

Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,3-  
15 dichloro-5-methoxypyridine (compound No. 193)

(a) A chloroform (60 ml) solution of 3.4 g (1 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-methoxypyridine (compound No. 244) was cooled with ice, 4.1 g (1.6 mmol) of m-chloroperbenzoic acid was added  
20 thereto, followed by stirring under cooling with ice for 2 hours, and stirring was further conducted at room temperature for 2 hours. 30 ml of a 0.5 mol/l aqueous sodium hydroxide solution was added to the mixture to terminate the reaction, the organic layer was dried over  
25 anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure to obtain 3.5 g (yield 85%) of 4-(2,3,4-trimethoxy-6-

methylbenzoyl)-3-chloro-5-methoxypyridine-N-oxide (m.p. 160-166°C).

(b) 5 ml of dimethylformamide was added to 2.5 ml of toluene, the mixture was cooled with ice, and 1.3 ml (1.4 mmol) of phosphorus oxychloride was dropwise added thereto. After the mixture was stirred under cooling with ice for 10 minutes, 2.5 g (0.7 mmol) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-chloro-5-methoxypyridine-N-oxide was added thereto. After the mixture was stirred under cooling with ice for 30 minutes, the temperature was recovered to room temperature, followed by stirring for 2 hours. 30 ml of water was added to the mixture to terminate the reaction, and ethyl acetate was added thereto for extraction. The organic layer was dried over anhydrous sodium sulfate, subjected to filtration and purified by silica gel column chromatography to obtain 2.0 g (yield 76%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2,3-dichloro-5-methoxypyridine (compound No. 193; m.p. 98-99°C).

#### SYNTHESIS EXAMPLE 15

##### Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2bromo-3-chloro-5-methoxypyridine (compound No. 245)

5 ml of dimethylformamide was added to 2.5 ml of toluene, the mixture was cooled with ice, and 0.7 g (0.2 mmol) of phosphorus oxybromide was dropwise added thereto. After the mixture was stirred under cooling with ice for 10 minutes, 0.42 g (0.1 mmol) of 4-(2,3,4-

trimethoxy-6-methylbenzoyl)-3-chloro-5-methoxypyridine-N-oxide obtained in Synthesis Example 14 (a) was added thereto. After the mixture was stirred under cooling with ice for 30 minutes, the temperature was recovered to room temperature, followed by stirring for 2 hours: 10 ml of water was added to the mixture to terminate the reaction, and ethyl acetate was added thereto for extraction. The organic layer was dried over anhydrous sodium sulfate, subjected to filtration and purified by silica gel column chromatography to obtain 0.32 g (yield 65%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-2bromo-3-chloro-5-methoxypyridine (compound No. 245; m.p. 97-99°C).

#### SYNTHESIS EXAMPLE 16

##### Synthesis of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-bromo-5-methylpyridine (compound No. 228)

(a) 57.0 ml (88.9 mmol) of n-butyllithium (1.56 M hexane solution) was dropwise added to a diethyl ether (110 ml) solution of 12.5 ml (89.2 mmol) of diisopropylamine at 0°C, followed by stirring for 60 minutes. The solution was cooled to -78°C, a toluene (80 ml) solution of 20 g (85 mmol) of 3,5-dibromopyridine was added thereto, followed by stirring for 5 minutes, and then a toluene 50 ml solution of 21.0 g (100 mmol) of 2,3,4-trimethoxy-6-methylbenzaldehyde was added thereto, followed by stirring for 2 hours. 50 ml of water was added to the mixture to terminate the reaction, the

aqueous layer was extracted with ethyl acetate, and the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 11.8 g (yield 31%) of (2,3,4-trimethoxy-6-methylphenyl)(3,5-dibromo-4-pyridyl)methanol (yellow oily substance).

(b) A tetrahydrofuran (15 mL) solution of 2.0 g (4.6 mmol) of (2,3,4-trimethoxy-6-methylphenyl)(3,5-dibromo-4-pyridyl)methanol obtained in step (a) was cooled to  $-78^{\circ}\text{C}$ , 6.0 mL (9.4 mmol) of n-butyllithium (1.56 M hexane solution) was dropwise added thereto, followed by stirring for 5 minutes, and 0.5 mL (8.0 mmol) of methyl iodide was added thereto, followed by stirring for 2.5 hours. 20 mL of water was added, and tetrahydrofuran was distilled off under reduced pressure. The aqueous layer was extracted with ethyl acetate, the organic layer was dried over anhydrous sodium sulfate and subjected to filtration, and the solvent was distilled off under reduced pressure. The crude product thus obtained was purified by silica gel column chromatography to obtain 0.44 g (yield 25%) of (2,3,4-trimethoxy-6-methylphenyl)(3-bromo-5-methyl-4-pyridyl)methanol.

(c) 3 g of manganese dioxide was added to a toluene (30 mL) solution of 0.43 g (1.1 mmol) of (2,3,4-trimethoxy-6-methylphenyl)(3-bromo-5-methyl-4-

pyridyl)methanol obtained in step (b), followed by stirring under reflux by heating for 2 hours. The mixture was subjected to filtration, the solvent was distilled off under reduced pressure, and the crude product thus obtained was purified by silica gel column chromatography to obtain 0.23 g (yield 54%) of 4-(2,3,4-trimethoxy-6-methylbenzoyl)-3-bromo-5-methylpyridine (compound No. 228; m.p. 88-93°C).

#### SYNTHESIS EXAMPLE FOR AN INTERMEDIATE

Now, Synthesis Example of 2,3,4-trimethoxy-6-methylbenzaldehyde to be used as an intermediate in the above Synthesis Examples 1, 3, 9, 11 and 16 is described below.

#### Synthesis of 2,3,4-trimethoxy-6-methylbenzaldehyde

A dry methylene chloride (100 ml) solution of 128 g (0.7 mol) of 3,4,5-trimethoxytoluene was dropwise added to a dry methylene chloride 500 ml solution of 112 g (0.84 mol) of aluminum chloride gradually under cooling with ice. The mixture was stirred at the same temperature for 45 minutes, a dry methylene chloride solution of 88.5 g (0.77 mol) of dichloromethyl methyl ether was dropwise added thereto gradually over a period of 2 hours. Stirring was conducted at the same temperature for 2 hours, and the mixture was gradually recovered to room temperature, followed by stirring at room temperature for one night. The reaction mixture was poured into 1 l of ice water, the methylene chloride phase

was separated, and the aqueous phase was extracted with 200 ml of methylene chloride twice. The extract and the methylene chloride phase were combined together, sequentially washed with 200 ml of water, 200 ml of a saturated aqueous sodium bicarbonate solution and 200 ml of a saturated aqueous sodium chloride solution and dried over anhydrous magnesium sulfate, and the solvent was distilled off under reduced pressure. A seed for a crystal was inoculated into the residue, and the resulting crystal was collected by filtration, washed with hexane and air dried to obtain 128 g of 2,3,4-trimethoxy-6-methylbenzaldehyde (m.p. 55-57°C).

Compounds produced by processes in accordance with Synthesis Examples 1 to 16 are shown in the following Tables 1 to 18.

Here, compounds represented by the formulae (I-1) to (I-9) in Tables are the following compounds. Further, in Tables, Me represents a methyl group, Et represents an ethyl group, Butyl represents a butyl group, i-Propyl represents an isopropyl group, Ph represents a phenyl group, Allyl represents an allyl group, c-Hexyl represents a cyclohexyl group, Benzyl represents a benzyl group, Propargyl represents a propargyl group, and Pentyl represents a pentyl group.

55

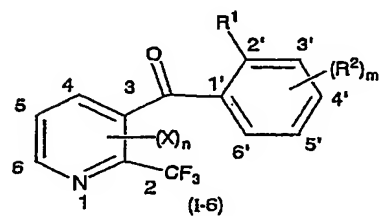
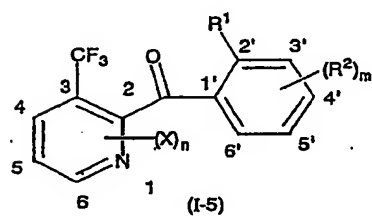
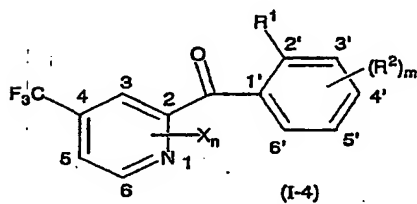
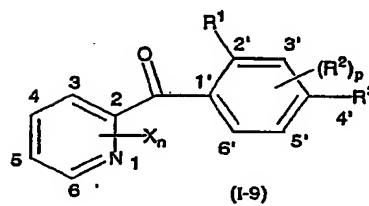
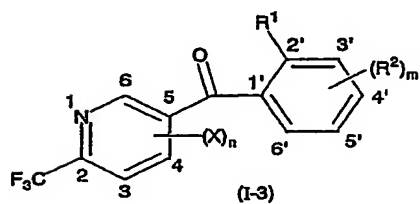
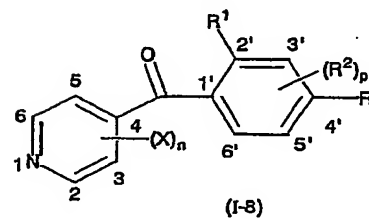
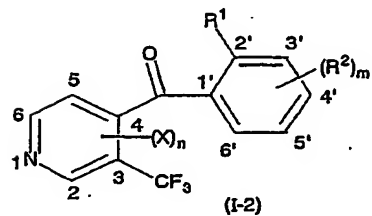
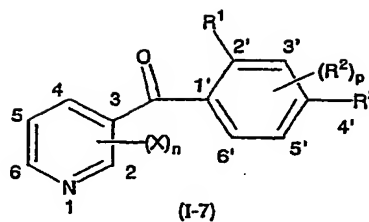
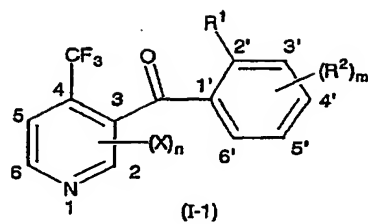


Table 1

Compounds represented by Formula (I-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
1	2-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	m.p.108-110°C
2	2-Cl, 6-Cl	Me	4'-MeO, 5'-Me	m.p.123-126°C
3	2-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.81-83°C
4	2-Cl, 6-Cl	Me	4'-MeO	Colorless oily substance
5	Not substituted	Me	4'-MeO, 5'-MeO	Yellow oily substance
6	Not substituted	Me	4'-MeO, 5'-Me	m.p.63-65°C
7	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.59-62°C
8	Not substituted	Me	4'-MeO	Pale yellow oily substance
9	2-Cl	Me	4'-MeO, 5'-MeO	m.p.82-86°C
10	2-Cl	Me	4'-MeO, 5'-Me	m.p.86-89°C
11	2-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.110-112°C
12	2-Cl	Me	4'-Me, 5'-Me, 6'-Me	m.p.88-95°C
13	2-Cl	Me	4',5'-(-OCH <sub>2</sub> O-)	m.p.119-122°C
14	2-Cl	Me	4'-MeO	Pale yellow oily substance
15	2-Cl	Et	4'-MeO, 5'-MeO, 6'-MeO	
16	2-Cl	i-Propyl	4'-MeO, 5'-MeO, 6'-MeO	
17	2-Cl	Me	3'-MeO, 4'-MeO, 5'-MeO, 6'-MeO	
18	2-Cl	Me	4'-MeO, 5'-EtO	m.p.89-90°C
19	2-Cl	Me	4'-MeO, 5'-i-Propyl-O-	Pale yellow oily substance
20	2-Cl	Me	4'-MeO, 5'-Allyl-O-	
21	2-Cl	Me	4'-MeO, 5'-Propargyl-O-	
22	2-Cl	Me	4'-MeO, 5'-CF <sub>3</sub> CH <sub>2</sub> O-	
23	2-Cl	Me	4'-MeO, 5'-c-Hexyl-O-	m.p.85-86°C

Table 2

Compounds represented by Formula (I-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
24	2-Cl	Me	4'-MeO, 5'-(CH <sub>3</sub> ) <sub>2</sub> N(CH <sub>2</sub> ) <sub>2</sub> O-	
25	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O-	
26	2-Cl	Me	4'-MeO, 5'-PhO-	
27	2-Cl	Me	4'-MeO, 5'-Benzyl-O-	m.p.116-117°C
28	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> COO-	
29	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> OCOO-	
30	2-MeO, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.93-94°C
31	2-Cl, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.85-87°C
32	2-MeO	Me	4'-MeO, 5'-MeO	m.p.125-127°C
33	2-i-Propyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
34	2-CF <sub>3</sub> CH <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.80-82°C
35	2-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
36	2-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
37	2-PhO-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
38	2-Benzyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
39	2-c-Hexyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
40	2-Allyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.85-86°C
41	2-Propargyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.121-124°C
42	2-(CH <sub>3</sub> ) <sub>2</sub> N(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
43	2-Cl, 5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.161-162°C
44	2-Cl, 5-Allyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
45	2-Cl, 5-Propargyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
46	2-Cl, 5-CH <sub>3</sub> O(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
47	2-Cl, 5-CH <sub>3</sub> (C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.127-129°C

Table 3

Compounds represented by Formula (I-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
48	2-Cl, 5-Et	Me	4'-MeO, 5'-MeO, 6'-MeO	
49	2-MeO, 5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
50	2-MeS	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
51	2-Me <sub>2</sub> N	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
52	2-Cl, 5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
53	2-CN	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 140-144°C
54	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 114-116°C
55	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 149-151°C
56	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 100-103°C
57	2-OH, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
58	2-Cl, 5-Me, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 101-104°C
59	2-Cl, 5-Allyl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
60	2-Cl, 5-Propargyl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
61	2-Cl, 5-CH <sub>3</sub> O(C=O)-, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 107-111°C
62	2-Cl, 5-CH <sub>3</sub> (C=O)-, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 109-112°C
63	2-Cl, 5-Et, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
64	2-Cl, 5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
65	5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
66	5-Allyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
67	5-Propargyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
68	5-CH <sub>3</sub> O(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
69	5-CH <sub>3</sub> (C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 110-113°C
70	5-Et	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 4

Compounds represented by Formula (I-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
71	5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
72	2-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO	m.p. 77-81°C
73	2-(6'-phenyl)-O-	Me	4'-MeO, 5'-MeO, 6'-O-(2-Pyridyl)	m.p. 183-189°C
74	2-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 81-82°C
75	2-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
76	2-MeS	Me	4'-MeO, 5'-MeO, 6'-OH	m.p. 98-102°C
77	2-OH	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 167-173°C
78	2-NH <sub>2</sub>	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 115-118°C
79	2-CH <sub>3</sub> NH	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 150-157°C
80	2-CH <sub>3</sub> COO	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
81	2-i-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-i-Propyl-O	Pale yellow oily substance
82	2-Cl, 6-Cl	Et	4'-MeO, 5'-MeO, 6'-MeO	m.p. 105-108°C
83	2-Cl	Me	4'-MeO, 5'-(4-MeO-Benzyl)O	m.p. 123-125°C
84	2-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 100-103°C
85	2-Me, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
86	2-Me, 5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 5

Compounds represented by Formula (I-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
87	2-PhO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 147-150°C
88	2-OH, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 6

Compounds represented by Formula (I-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
89	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO	m.p.120-125°C
90	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.106-109°C
91	Not substituted	Me	4'-MeO, 5'-MeO	m.p.98-101°C
92	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.104-107°C
93	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.130-134°C
94	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO	m.p.151-156°C
95	2-Br, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
96	2-MeS, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
97	2-CN, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
98	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	m.p.139-141°C
99	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.113-115°C
100	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	m.p.94-97°C
101	5-Cl	Me	4'-MeO, 5'-MeO	m.p.90-91°C
102	5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.75-77°C
103	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.72-74°C
104	5-Cl, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.143-146°C
105	5-Cl, 6-MeO	Me	4'-MeO, 5'-MeO	m.p.112-115°C
106	5-Cl, 6-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.82-84°C
107	6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
108	5-Cl, 6-n-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
109	6-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
110	5-Cl, 6-n-Butyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
111	6-n-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
112	6-n-Butyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance

Table 7

Compounds represented by Formula (I-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
113	5-Cl, 6-Propargyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.131-133°C
114	5-Cl, 6-n-Pentyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
115	5-Cl, 6-OH	Me	4'-MeO, 5'-MeO, 6'-MeO	152-154°C
116	6-n-Pentyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
117	5-Cl, 6-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.62-64°C
118	5-Cl, 6-Allyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	Pale yellow oily substance
119	5-Cl, 6-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.78-80°C
120	2-MeO, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.135-139°C
121	2-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.97-100°C
122	5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.103-106°C
123	2-Cl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.117-119°C
124	2-Br, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.145-147°C
125	2-Me, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
126	2-Et, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
127	2-n-Propyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
128	2-Allyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
129	2-Propargyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
130	2-EtO, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.110-112°C
131	2-CN, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.117-120°C
132	2-MeS, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.154-159°C
133	5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.98-105°C
134	5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	
135	5-F	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 8

Compound represented by Formula (I-3)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
136	6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Yellow oily substance

Table 9

Compound represented by Formula (I-4)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
137	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 10

Compound represented by Formula (I-5)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
138	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 109-111°C

Table 11

Compounds represented by Formula (I-6)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
139	4-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
140	4-Me, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
141	4-Me, 5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 12

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
142	(I-7)	2-MeO	Me	5'-MeO	MeO	m.p. 90-91°C
143	(I-7)	2-Cl, 4-Cl	Me	5'-MeO	PhO	
144	(I-7)	2-Cl, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
145	(I-7)	2-Cl, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
146	(I-7)	2-MeO, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
147	(I-7)	2-F, 4-F	Me	5'-MeO, 6'-MeO	MeO	
148	(I-7)	2-F, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
149	(I-7)	2-MeO, 4-F	Me	5'-MeO, 6'-MeO	MeO	
150	(I-7)	2-Cl, 4-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
151	(I-7)	2-Me, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
152	(I-7)	2-Me, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
153	(I-7)	2-Me, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
154	(I-7)	2-Me, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
155	(I-7)	2-Me, 4-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
156	(I-7)	2-Me, 4-MeO, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
157	(I-7)	2-MeO, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
158	(I-7)	2-MeO, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
159	(I-7)	2-MeO, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
160	(I-7)	2-Me, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
161	(I-7)	2-Me, 4-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
162	(I-7)	2-Me, 4-Cl, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
163	(I-7)	2-Cl, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
164	(I-7)	2-Cl, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
165	(I-7)	2-Cl, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
166	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 13

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
167	(I-7)	2-Cl, 4-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	m.p. 117-118°C
168	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Me	Me	5'-MeO, 6'-MeO	MeO	
169	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Et	Me	5'-MeO, 6'-MeO	MeO	
170	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Allyl	Me	5'-MeO, 6'-MeO	MeO	
171	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-n-Propyl	Me	5'-MeO, 6'-MeO	MeO	
172	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Propargyl	Me	5'-MeO, 6'-MeO	MeO	
173	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Me, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
174	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Et, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
175	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Allyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
176	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-n-Propyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
177	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Propargyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
178	(I-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
179	(I-7)	2-CF <sub>3</sub> , 5-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
180	(I-7)	2-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
181	(I-7)	2-CF <sub>3</sub> , 4-Me, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
182	(I-7)	2-CF <sub>3</sub> , 4-Et, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
183	(I-7)	2-CF <sub>3</sub> , 4-Allyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
184	(I-7)	2-CF <sub>3</sub> , 4-n-propyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
185	(I-7)	2-CF <sub>3</sub> , 4-propargyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
186	(I-8)	2-Cl, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 60-61°C
187	(I-8)	2-MeO, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 128-134°C

Table 14

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
188	(I-8)	2-EtO, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
189	(I-8)	2-MeO, 3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
190	(I-8)	2-MeO, 3-MeO	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
191	(I-8)	3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 109-111°C
192	(I-8)	3-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 90-94°C
193	(I-8)	2-Cl, 3-Cl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	m.p. 98-99°C
194	(I-8)	2-Cl, 3-Cl, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	m.p. 110-114°C
195	(I-8)	2-Cl, 3-MeO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
196	(I-8)	2-Cl, 3-EtO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
197	(I-8)	3-MeO	Me	5'-MeO, 6'-MeO	MeO	
198	(I-8)	3-EtO	Me	5'-MeO, 6'-MeO	MeO	
199	(I-8)	2-Cl, 3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 80-86°C
200	(I-8)	2-Cl, 3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
201	(I-8)	3-Br	Me	5'-MeO, 6'-MeO	MeO	m.p. 106-107°C
202	(I-8)	3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	m.p. 108-110°C
203	(I-8)	3-Br, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
204	(I-8)	2-F, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
205	(I-8)	2-MeO, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
206	(I-8)	2-EtO, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
207	(I-8)	2-MeO, 3-MeO, 5-F	Me	5'-MeO, 6'-MeO	MeO	
208	(I-8)	3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
209	(I-8)	3-F	Me	5'-MeO, 6'-MeO	MeO	
210	(I-8)	3-Me, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance

Table 15

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
211	(I-8)	2-Cl, 3-Me, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
212	(I-8)	2-Br, 3-Me, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
213	(I-8)	3-Me, 5-Me	Me	5'-MeO, 6'-MeO	MeO	m.p. 117-122°C
214	(I-8)	2-Cl, 3-Me, 5-Me	Me	5'-MeO, 6'-MeO	MeO	
215	(I-8)	2-Br, 3-Me, 5-Me	Me	5'-MeO, 6'-MeO	MeO	
216	(I-8)	3-Et, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
217	(I-8)	3-Allyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
218	(I-8)	3-n-Propyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
219	(I-8)	3-Propargyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
220	(I-8)	2-Cl, 3-Et, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
221	(I-8)	2-Cl, 3-Allyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
222	(I-8)	2-Cl, 3-n-Propyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
223	(I-8)	2-Cl, 3-Propargyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
224	(I-8)	2-Br, 3-Et, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
225	(I-8)	2-Br, 3-Allyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
226	(I-8)	2-Br, 3-n-Propyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
227	(I-8)	2-Br, 3-Propargyl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
228	(I-8)	3-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	m.p. 88-93°C
229	(I-8)	3-Et, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
230	(I-8)	3-Allyl, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
231	(I-8)	3-n-Propyl, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
232	(I-8)	3-Propargyl, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
233	(I-8)	3-Me, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
234	(I-8)	3-Et, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 16

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
235	(I-8)	3-Allyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
236	(I-8)	3-n-Propyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
237	(I-8)	3-Propargyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
238	(I-8)	3-Me, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
239	(I-8)	3-Et, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
240	(I-8)	3-Allyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
241	(I-8)	3-n-Propyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
242	(I-8)	3-Propargyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
243	(I-8)	3-MeO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Red oily substance
244	(I-8)	3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
245	(I-8)	2-Br, 3-Cl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	m.p. 97-99°C
246	(I-8)	2-Br, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
247	(I-8)	2-Cl, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
248	(I-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
249	(I-8)	2-Cl, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	Yellow oily substance
250	(I-8)	2-Br, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	Yellow oily substance
251	(I-8)	3-EtO, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	m.p. 106-109°C
252	(I-8)	3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 98.5-99.5°C
253	(I-8)	2-Br, 3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
254	(I-8)	2-Br, 3-Cl, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	m.p. 113-115°C
255	(I-8)	2-Cl, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
256	(I-8)	2-Br, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
257	(I-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
258	(I-8)	2-Cl, 3-EtO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 17

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
259	(I-8)	2-Br, 3-EtO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
260	(I-8)	2-Cl, 3-EtO, 5-Cl, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
261	(I-8)	2-Br, 3-EtO, 5-Cl, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
262	(I-8)	2-F, 3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	m.p. 85-87°C
263	(I-8)	2-Br, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
264	(I-8)	2-F, 3-Me, 5-F	Me	5'-MeO, 6'-MeO	MeO	
265	(I-8)	2-Br, 3-F, 5-F, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
266	(I-8)	2-Cl, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
267	(I-8)	2-Br, 3-Br, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
268	(I-8)	2-Cl, 3-Cl, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
269	(I-8)	3-Br, 5-F	Me	5'-MeO, 6'-MeO	MeO	
270	(I-8)	2-Br, 3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	
271	(I-8)	3-F, 5-CH <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
272	(I-8)	3-Cl, 5-CH <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	m.p. 84-88°C
273	(I-8)	3-F, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
274	(I-8)	2-Cl, 3-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	m.p. 85-88°C
275	(I-8)	3-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
276	(I-8)	3-CF <sub>3</sub> , 5-Me, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
277	(I-8)	3-CF <sub>3</sub> , 5-Et, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
278	(I-8)	3-CF <sub>3</sub> , 5-Allyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
279	(I-8)	3-CF <sub>3</sub> , 5-n-Propyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
280	(I-8)	3-CF <sub>3</sub> , 5-Propargyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
281	(I-8)	2-Cl, 3-CF <sub>3</sub> , 5-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
282	(I-8)	2-Cl, 3-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	

Table 18

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
283	(I-8)	3-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
284	(I-9)	3-Cl, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 144-147°C
285	(I-9)	3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	
286	(I-9)	3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	Pale yellow oily substance
287	(I-1)	2-MeO	Me	5'-C-Hexyl-O	MeO	m.p. 97-100°C
288	(I-8)	2-Me, 3-Cl, 6-Cl	Me	5'-MeO	MeO	m.p. 111-113°C
289	(I-8)	2-Me, 3-Cl, 6-Cl	Et	5'-MeO	MeO	m.p. 88-94°C
290	(I-8)	2-Me, 3-Cl	Me	5'-MeO	MeO	m.p. 117-118°C
291	(I-8)	2-Cl, 3-Br, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Brown oily substance
292	(I-8)	2-Br, 3-Br, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	Yellow oily substance

Compounds represented by the formula (X) to be used as an intermediate, produced by processes in accordance with Synthesis Examples 1, 3, 5, 6, 8, 9, 11 and 16, are shown in the following Tables 19 to 36.

Here, compounds represented by the general formulae (X-1) to (X-9) in Tables are the following compounds.

Further, in Tables, Me represents a methyl group, Et represents an ethyl group, Butyl represents a butyl group, i-Propyl represents an isopropyl group, Ph represents a phenyl group, Allyl represents an allyl group, c-Hexyl represents a cyclohexyl group, Benzyl represents a benzyl group, Propargyl represents a propargyl group, and Pentyl represents a pentyl group.

71

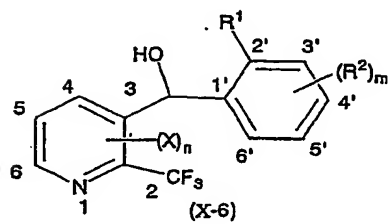
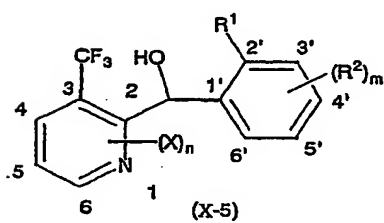
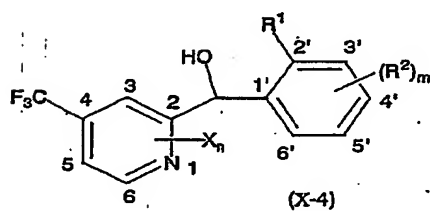
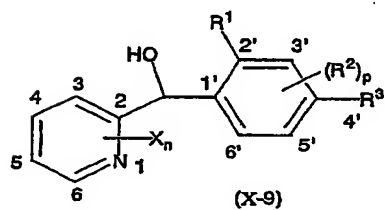
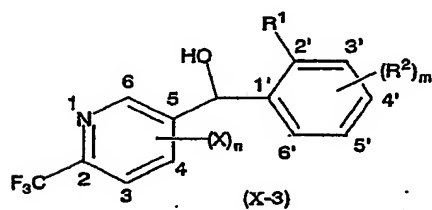
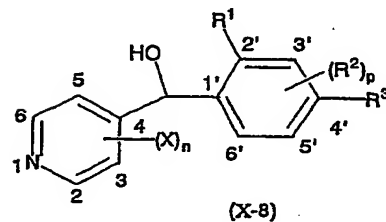
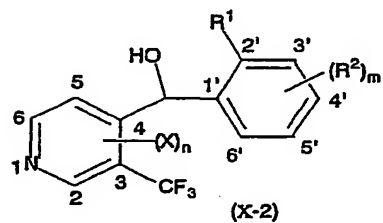
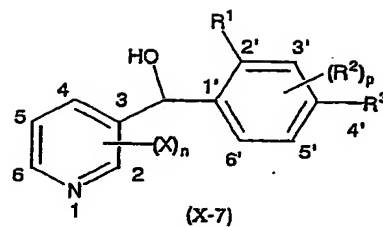
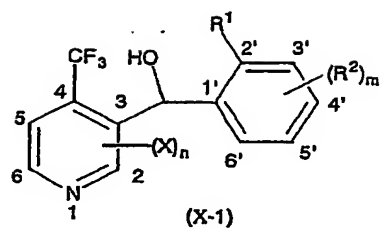


Table 19

Compounds represented by Formula (X-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
1	2-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	Viscous substance
2	2-Cl, 6-Cl	Me	4'-MeO, 5'-Me	Viscous substance
3	2-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance
4	2-Cl, 6-Cl	Me	4'-MeO	Viscous substance
5	Not substituted	Me	4'-MeO, 5'-MeO	
6	Not substituted	Me	4'-MeO, 5'-Me	
7	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 132-135°C
8	Not substituted	Me	4'-MeO	
9	2-Cl	Me	4'-MeO, 5'-MeO	
10	2-Cl	Me	4'-MeO, 5'-Me	
11	2-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance
12	2-Cl	Me	4'-Me, 5'-Me, 6'-Me	m.p. 125-127°C
13	2-Cl	Me	4', 5'-(-OCH <sub>2</sub> O-)	m.p. 127-130°C
14	2-Cl	Me	4'-MeO	
15	2-Cl	Et	4'-MeO, 5'-MeO, 6'-MeO	
16	2-Cl	i-Propyl	4'-MeO, 5'-MeO, 6'-MeO	
17	2-Cl	Me	3'-MeO, 4'-MeO, 5'-MeO, 6'-MeO	
18	2-Cl	Me	4'-MeO, 5'-EtO	Viscous substance
19	2-Cl	Me	4'-MeO, 5'-i-Propyl-O-	Viscous substance
20	2-Cl	Me	4'-MeO, 5'-Allyl-O-	
21	2-Cl	Me	4'-MeO, 5'-Propargyl-O-	
22	2-Cl	Me	4'-MeO, 5'-CF <sub>3</sub> CH <sub>2</sub> O-	
23	2-Cl	Me	4'-MeO, 5'-c-Hexyl-O-	Viscous substance

Table 20

Compounds represented by Formula (X-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
24	2-Cl	Me	4'-MeO, 5'-(CH <sub>3</sub> ) <sub>2</sub> N(CH <sub>2</sub> ) <sub>2</sub> O-	
25	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O-	
26	2-Cl	Me	4'-MeO, 5'-PhO-	
27	2-Cl	Me	4'-MeO, 5'-Benzyl-O-	Viscous substance
28	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> COO-	
29	2-Cl	Me	4'-MeO, 5'-CH <sub>3</sub> OCO-	
30	2-MeO, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
31	2-Cl, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
32	2-MeO	Me	4'-MeO, 5'-MeO	
33	2-i-Propyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
34	2-CF <sub>3</sub> CH <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
35	2-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
36	2-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
37	2-PhO-	Me	4'-MeO, 5'-MeO, 6'-MeO	
38	2-Benzyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
39	2-c-Hexyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
40	2-Allyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
41	2-Propargyl-O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
42	2-(CH <sub>3</sub> ) <sub>2</sub> N(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO, 6'-MeO	
43	2-Cl, 5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
44	2-Cl, 5-Allyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
45	2-Cl, 5-Propargyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
46	2-Cl, 5-CH <sub>3</sub> O(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
47	2-Cl, 5-CH <sub>3</sub> (C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 21

Compounds represented by Formula (X-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
48	2-Cl, 5-Et	Me	4'-MeO, 5'-MeO, 6'-MeO	
49	2-MeO, 5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
50	2-MeS	Me	4'-MeO, 5'-MeO, 6'-MeO	
51	2-Me <sub>2</sub> N	Me	4'-MeO, 5'-MeO, 6'-MeO	
52	2-Cl, 5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
53	2-CN	Me	4'-MeO, 5'-MeO, 6'-MeO	
54	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance
55	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
56	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
57	2-OH, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
58	2-Cl, 5-Me, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance
59	2-Cl, 5-Allyl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance
60	2-Cl, 5-Propargyl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
61	2-Cl, 5-CH <sub>3</sub> O(C=O)-, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 133-135°C
62	2-Cl, 5-CH <sub>3</sub> CH(OH)-, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 141-158°C
63	2-Cl, 5-Et, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
64	2-Cl, 5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
65	5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
66	5-Allyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
67	5-Propargyl	Me	4'-MeO, 5'-MeO, 6'-MeO	
68	5-CH <sub>3</sub> O(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
69	5-CH <sub>3</sub> (C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
70	5-Et	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 22

Compounds represented by Formula (X-1)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
71	5-(CH <sub>3</sub> ) <sub>2</sub> N(C=O)-	Me	4'-MeO, 5'-MeO, 6'-MeO	
72	2-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O-	Me	4'-MeO, 5'-MeO	
73	2-(6'-phenyl)-O-	Me	4'-MeO, 5'-MeO, 6'-O-(2-Pyridyl)	
74	2-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
75	2-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	
76	2-MeS	Me	4'-MeO, 5'-MeO, 6'-OH	
77	2-OH	Me	4'-MeO, 5'-MeO, 6'-MeO	
78	2-NH <sub>2</sub>	Me	4'-MeO, 5'-MeO, 6'-MeO	
79	2-CH <sub>3</sub> NH	Me	4'-MeO, 5'-MeO, 6'-MeO	
80	2-CH <sub>3</sub> COO	Me	4'-MeO, 5'-MeO, 6'-MeO	
81	2-i-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-i-Propyl-O	
82	2-Cl, 6-Cl	Et	4'-MeO, 5'-MeO, 6'-MeO	
83	2-Cl	Me	4'-MeO, 5'-(4-MeO-Benzyl)O	Viscous substance
84	2-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
85	2-Me, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
86	2-Me, 5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 23

Compounds represented by Formula (X-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
87	2-PhO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
88	2-OH, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 24

Compounds represented by Formula (X-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
89	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO	m.p.134-136°C
90	2-Cl, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.162-165°C
91	Not substituted	Me	4'-MeO, 5'-MeO	
92	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.101-106°C
93	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
94	2-MeO, 5-Cl	Me	4'-MeO, 5'-MeO	
95	2-Br, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
96	2-MeS, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
97	2-CN, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
98	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	m.p.156-158°C
99	2-Cl, 5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.131-135°C
100	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO	Viscous substance
101	5-Cl	Me	4'-MeO, 5'-MeO	
102	5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.102-105°C
103	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p.95-98°C
104	5-Cl, 6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
105	5-Cl, 6-MeO	Me	4'-MeO, 5'-MeO	
106	5-Cl, 6-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	
107	6-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
108	5-Cl, 6-n-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
109	6-EtO	Me	4'-MeO, 5'-MeO, 6'-MeO	
110	5-Cl, 6-n-Butyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
111	6-n-Propyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
112	6-n-Butyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 25

Compounds represented by Formula (X-2)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
113	5-Cl, 6-Propargyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
114	5-Cl, 6-n-Pentyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
115	5-Cl, 6-OH	Me	4'-MeO, 5'-MeO, 6'-MeO	
116	6-n-Pentyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
117	5-Cl, 6-CH <sub>3</sub> S(CH <sub>2</sub> ) <sub>2</sub> O	Me	4'-MeO, 5'-MeO, 6'-MeO	
118	5-Cl, 6-Allyl-O	Me	4'-MeO, 5'-MeO, 6'-MeO	
119	5-Cl, 6-CH <sub>3</sub> O(CH <sub>2</sub> ) <sub>2</sub> O	Me	4'-MeO, 5'-MeO, 6'-MeO	
120	2-MeO, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
121	2-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
122	5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
123	2-Cl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
124	2-Br, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
125	2-Me, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
126	2-Et, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
127	2-n-Propyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
128	2-Allyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
129	2-Propargyl, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
130	2-EtO, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
131	2-CN, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
132	2-MeS, 5-MeO	Me	4'-MeO, 5'-MeO, 6'-MeO	
133	5-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
134	5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	
135	5-F	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 26

Compound represented by Formula (X-3)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
136	6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	Viscous substance

Table 27

Compound represented by Formula (X-4)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
137	Not substituted	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 28

Compound represented by Formula (X-5)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
138	5-Cl, 6-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	m.p. 71-73°C

Table 29

Compounds represented by Formula (X-6)				
No.	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>m</sub>	Physical properties
139	4-Me	Me	4'-MeO, 5'-MeO, 6'-MeO	
140	4-Me, 5-Cl	Me	4'-MeO, 5'-MeO, 6'-MeO	
141	4-Me, 5-Br	Me	4'-MeO, 5'-MeO, 6'-MeO	

Table 30

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
142	(X-7)	2-MeO	Me	5'-MeO	MeO	Viscous substance
143	(X-7)	2-Cl, 4-Cl	Me	5'-MeO	PhO	
144	(X-7)	2-Cl, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
145	(X-7)	2-Cl, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
146	(X-7)	2-MeO, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
147	(X-7)	2-F, 4-F	Me	5'-MeO, 6'-MeO	MeO	
148	(X-7)	2-F, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
149	(X-7)	2-MeO, 4-F	Me	5'-MeO, 6'-MeO	MeO	
150	(X-7)	2-Cl, 4-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
151	(X-7)	2-Me, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
152	(X-7)	2-Me, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
153	(X-7)	2-Me, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
154	(X-7)	2-Me, 4-MeO	Me	5'-MeO, 6'-MeO	MeO	
155	(X-7)	2-Me, 4-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
156	(X-7)	2-Me, 4-MeO, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
157	(X-7)	2-MeO, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
158	(X-7)	2-MeO, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
159	(X-7)	2-MeO, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
160	(X-7)	2-Me, 4-Cl	Me	5'-MeO, 6'-MeO	MeO	
161	(X-7)	2-Me, 4-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
162	(X-7)	2-Me, 4-Cl, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
163	(X-7)	2-Cl, 4-Me	Me	5'-MeO, 6'-MeO	MeO	
164	(X-7)	2-Cl, 4-Me, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
165	(X-7)	2-Cl, 4-Me, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
166	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 31

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
167	(X-7)	2-Cl, 4-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	Viscous substance
168	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Me	Me	5'-MeO, 6'-MeO	MeO	
169	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Et	Me	5'-MeO, 6'-MeO	MeO	
170	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Allyl	Me	5'-MeO, 6'-MeO	MeO	
171	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-n-Propyl	Me	5'-MeO, 6'-MeO	MeO	
172	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Propargyl	Me	5'-MeO, 6'-MeO	MeO	
173	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Me, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
174	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Et, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
175	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Allyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
176	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-n-Propyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
177	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub> , 5-Propargyl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
178	(X-7)	2-CF <sub>3</sub> , 4-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
179	(X-7)	2-CF <sub>3</sub> , 5-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
180	(X-7)	2-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
181	(X-7)	2-CF <sub>3</sub> , 4-Me, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
182	(X-7)	2-CF <sub>3</sub> , 4-Et, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
183	(X-7)	2-CF <sub>3</sub> , 4-Allyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
184	(X-7)	2-CF <sub>3</sub> , 4-n-propyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
185	(X-7)	2-CF <sub>3</sub> , 4-propargyl, 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
186	(X-8)	2-Cl, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	Amorphous
187	(X-8)	2-MeO, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 32

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
188	(X-8)	2-EtO, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
189	(X-8)	2-MeO, 3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
190	(X-8)	2-MeO, 3-MeO	Me	5'-MeO, 6'-MeO	MeO	
191	(X-8)	3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 136-140°C
192	(X-8)	3-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 160-162°C
193	(X-8)	2-Cl, 3-Cl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
194	(X-8)	2-Cl, 3-Cl, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
195	(X-8)	2-Cl, 3-MeO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
196	(X-8)	2-Cl, 3-EtO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
197	(X-8)	3-MeO	Me	5'-MeO, 6'-MeO	MeO	
198	(X-8)	3-EtO	Me	5'-MeO, 6'-MeO	MeO	
199	(X-8)	2-Cl, 3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
200	(X-8)	2-Cl, 3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
201	(X-8)	3-Br	Me	5'-MeO, 6'-MeO	MeO	m.p. 168-169°C
202	(X-8)	3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	Viscous substance
203	(X-8)	3-Br, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	m.p. 90-93°C
204	(X-8)	2-F, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
205	(X-8)	2-MeO, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
206	(X-8)	2-EtO, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
207	(X-8)	2-MeO, 3-MeO, 5-F	Me	5'-MeO, 6'-MeO	MeO	
208	(X-8)	3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
209	(X-8)	3-F	Me	5'-MeO, 6'-MeO	MeO	
210	(X-8)	3-Me, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	

Table 34

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
235	(X-8)	3-Allyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
236	(X-8)	3-n-Propyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
237	(X-8)	3-Propargyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
238	(X-8)	3-Me, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
239	(X-8)	3-Et, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
240	(X-8)	3-Allyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
241	(X-8)	3-n-Propyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
242	(X-8)	3-Propargyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
243	(X-8)	3-MeO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
244	(X-8)	3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
245	(X-8)	2-Br, 3-Cl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
246	(X-8)	2-Br, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
247	(X-8)	2-Cl, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
248	(X-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
249	(X-8)	2-Cl, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
250	(X-8)	2-Br, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
251	(X-8)	3-EtO, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
252	(X-8)	3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
253	(X-8)	2-Br, 3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
254	(X-8)	2-Br, 3-Cl, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
255	(X-8)	2-Cl, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
256	(X-8)	2-Br, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
257	(X-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
258	(X-8)	2-Cl, 3-EtO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 34

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
235	(X-8)	3-Allyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
236	(X-8)	3-n-Propyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
237	(X-8)	3-Propargyl, 5-Br, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
238	(X-8)	3-Me, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
239	(X-8)	3-Et, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
240	(X-8)	3-Allyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
241	(X-8)	3-n-Propyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
242	(X-8)	3-Propargyl, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
243	(X-8)	3-MeO, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
244	(X-8)	3-MeO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
245	(X-8)	2-Br, 3-Cl, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
246	(X-8)	2-Br, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
247	(X-8)	2-Cl, 3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	
248	(X-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
249	(X-8)	2-Cl, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
250	(X-8)	2-Br, 3-MeO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
251	(X-8)	3-EtO, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
252	(X-8)	3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
253	(X-8)	2-Br, 3-EtO, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
254	(X-8)	2-Br, 3-Cl, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
255	(X-8)	2-Cl, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
256	(X-8)	2-Br, 3-Br, 5-EtO	Me	5'-MeO, 6'-MeO	MeO	
257	(X-8)	2-Br, 3-Cl, 5-Cl	Me	5'-MeO, 6'-MeO	MeO	
258	(X-8)	2-Cl, 3-EtO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	

Table 35

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
259	(X-8)	2-Br, 3-EtO, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
260	(X-8)	2-Cl, 3-EtO, 5-Cl, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
261	(X-8)	2-Br, 3-EtO, 5-Cl, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
262	(X-8)	2-F, 3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	Viscous substance
263	(X-8)	2-Br, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
264	(X-8)	2-F, 3-Me, 5-F	Me	5'-MeO, 6'-MeO	MeO	
265	(X-8)	2-Br, 3-F, 5-F, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
266	(X-8)	2-Cl, 3-F, 5-F	Me	5'-MeO, 6'-MeO	MeO	
267	(X-8)	2-Br, 3-Br, 5-Br, 6-Br	Me	5'-MeO, 6'-MeO	MeO	
268	(X-8)	2-Cl, 3-Cl, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
269	(X-8)	3-Br, 5-F	Me	5'-MeO, 6'-MeO	MeO	
270	(X-8)	2-Br, 3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	
271	(X-8)	3-F, 5-CH <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
272	(X-8)	3-Cl, 5-CH <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
273	(X-8)	3-F, 5-MeO	Me	5'-MeO, 6'-MeO	MeO	
274	(X-8)	2-Cl, 3-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	m.p. 154-158°C
275	(X-8)	3-CF <sub>3</sub> , 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
276	(X-8)	3-CF <sub>3</sub> , 5-Me, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
277	(X-8)	3-CF <sub>3</sub> , 5-Et, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
278	(X-8)	3-CF <sub>3</sub> , 5-Allyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
279	(X-8)	3-CF <sub>3</sub> , 5-n-Propyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
280	(X-8)	3-CF <sub>3</sub> , 5-Propargyl, 6-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
281	(X-8)	2-Cl, 3-CF <sub>3</sub> , 5-CF <sub>3</sub> , 6-Cl	Me	5'-MeO, 6'-MeO	MeO	
282	(X-8)	2-Cl, 3-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	

Table 36

No.	Formula	(X) <sub>n</sub>	R <sup>1</sup>	(R <sup>2</sup> ) <sub>p</sub>	R <sup>3</sup>	Physical properties
283	(X-8)	3-CF <sub>3</sub> , 5-CF <sub>3</sub>	Me	5'-MeO, 6'-MeO	MeO	
284	(X-9)	3-Cl, 5-Cl, 6-Cl	Me	5'-MeO, 6'-MeO	MeO	m.p. 97-99°C
285	(X-9)	3-F, 5-F, 6-F	Me	5'-MeO, 6'-MeO	MeO	
286	(X-9)	3-Br, 5-Br	Me	5'-MeO, 6'-MeO	MeO	m.p. 114-117°C

The benzoylpyridine derivative represented by the formula (I) or its salt is useful as an active ingredient for a fungicide, particularly as an active ingredient for an agricultural and horticultural fungicide. As the

5 agricultural and horticultural fungicide, it is effective for controlling diseases such as blast, brown spot or sheath blight of rice (Oryza sativa); powdery mildew, scab, rust, snow mold, loose smut, eyespot, leaf spot or glume blotch of barley (Hordeum vulgare); melanose or

10 scab of citrus (Citrus); blossom blight, powdery mildew, Alternaria leaf spot or scab of apple (Malus pumila); scab or black spot of pear (Pyrus serotina, Pyrus ussuriensis, Pyrus communis); brown rot, scab or Fomitopsis rot of peach (Prunus persica); Anthracnose, ripe rot, powdery

15 mildew or downy mildew of grape (Vitis vinifera); anthracnose or circular leaf spot of Japanese persimmon (Diospyros kaki); anthracnose, powdery mildew, gummy stem blight or downy mildew of cucurbit (Cucumis melo); early blight, leaf mold or late blight of tomato (Lycopersicon

20 esculentum); leaf blight of cress (Brassica sp., Raphanus sp., etc); early blight or late blight of potato (Solanum tuberosum); powdery mildew of strawberry (Fragaria chiloensis); gray mold or stem rot of various crops. It shows an excellent controlling effect particularly on

25 powdery mildew of barley and vegetables and rice blast. Further, it is also effective for controlling soil-borne diseases caused by phytopathogenic fungi such as

Fusarium, Pythium, Rhizoctonia, Verticillium and Plasmodiophora.

The compound of the present invention may be used in combination with an agricultural adjuvant to formulate various preparations of the fungicide containing the compound, such as a dust, granules, a granular wettable powder, a wettable powder, an aqueous suspension, an oil suspension, a water soluble powder, an emulsifiable concentrate, an aqueous solution, a paste, an aerosol or a microdose dusting powder. The compound of the present invention may be formed into any preparation which is usually used in the agricultural and horticultural field so long as the purpose of the present invention is met. The adjuvant to be used for preparation may, for example, be a solid carrier such as diatomaceous earth, hydrated lime, calcium carbonate, talc, white carbon, kaolin, bentonite, a mixture of kaolinite and sericite, clay, sodium carbonate, sodium bicarbonate, glauher's salt, zeolite or starch; a solvent such as water, toluene, xylene, solvent naphtha, dioxane, acetone, isophorone, methyl isobutyl ketone, chlorobenzene, cyclohexane, dimethylsulfoxide, dimethylformamide, dimethylacetamide, N-methyl-2-pyrrolidone or an alcohol; an anionic surfactant or spreading agent such as a fatty acid salt, a benzoate, an alkyl sulfosuccinate, a dialkyl sulfosuccinate, a polycarboxylate, an alkyl sulfuric ester salt, an alkyl sulfate, an alkyl aryl sulfate, an

alkyl diglycol ether sulfate, an alcohol sulfuric ester salt, an alkyl sulfonate, an alkyl aryl sulfonate, an aryl sulfonate, a lignin sulfonate, an alkyl diphenyl ether disulfonate, a polystyrene sulfonate, an alkyl  
5 phosphoric ester salt, an alkyl aryl phosphate, a styryl aryl phosphate, a polyoxyethylene alkyl ether sulfuric ester salt, a polyoxyethylene alkyl aryl ether sulfate, a polyoxyethylene alkyl aryl ether sulfuric ester salt, a polyoxyethylene alkyl ether phosphate, a polyoxyethylene  
10 alkyl aryl phosphoric ester salt or a salt of a naphthalene sulfonic acid formalin condensate; a non-ionic surfactant or spreading agent such as a sorbitan fatty acid ester, a glycerol fatty acid ester, a fatty acid polyglyceride, a fatty acid alcohol polyglycol  
15 ether, an acetylene glycol, an acetylene alcohol, an oxyalkylene block polymer, a polyoxyethylene alkyl ether, a polyoxyethylene alkyl aryl ether, a polyoxyethylene styryl aryl ether, a polyoxyethylene glycol alkyl ether, a polyoxyethylene fatty acid ester, a polyoxyethylene  
20 sorbitan fatty acid ester, a polyoxyethylene glycerol fatty acid ester, a polyoxyethylene hardened castor oil or a polyoxypropylene fatty acid ester; vegetable oil or mineral oil such as olive oil, kapok oil, castor oil, palm oil, camellia oil, coconut oil, sesame oil, corn  
25 oil, rice bran oil, peanut oil, cotton oil, soy bean oil, rape oil, linseed oil, tung oil or liquid paraffin. A known adjuvant may be selected from adjuvants which are

known in the agricultural and horticultural field within a range of not departing from the object of the present invention. Further, an adjuvant which is usually used may also be employed, such as a bulking agent, a  
5 thickener, an anti-settling agent, a freeze proofing agent, a dispersion stabilizer, a crop injury-reducing agent or a mildewproofing agent. The blending proportion of the compound of the present invention to the adjuvant is generally from 0.005:99.995 to 95:5, preferably from  
10 0.2:99.8 to 90:10. These formulations can be practically used either as they are or after they are diluted with a diluent such as water to predetermined concentrations and a spreading agent is added thereto as the case requires.

The concentration of the compound of the present  
15 invention varies depending upon the crop plant as the object, the way of application, the form of preparation or the dose, and hence cannot be generically determined. However, in the case of foliage treatment, the concentration of the compound as the active ingredient is  
20 generally from 0.1 to 10,000 ppm, preferably from 1 to 2,000 ppm. In the case of soil treatment, it is generally from 10 to 100,000 g/ha, preferably from 200 to 20,000 g/ha.

The preparation fungicide containing the compound of  
25 the present invention or a diluted product thereof can be applied by an application method which is commonly used, such as spreading (spreading, spraying, misting,

atomizing, grain diffusing or application on water), soil application (such as mixing or irrigation) or surface application (such as coating, dust coating or covering). Further, it may be applied also by so-called ultra low  
5 volume. By this method, the preparation can contain 100% of the active ingredient.

The fungicide of the present invention may be mixed or used together with e.g. another agricultural chemical such as an insecticide, a miticide, a nematocide, a  
10 fungicide, an antiviral agent, an attractant, an herbicide or a plant growth regulator. In such a case, a still more excellent effect may be obtained in some cases.

Examples of the active ingredient compound (generic  
15 name; including compounds which are under application) of the insecticide, miticide or nematocide i.e. a pesticide of the above other agricultural chemicals, include organic phosphate type compounds such as Profenofos, Dichlorvos, Fenamiphos, Fenitrothion, EPN, Diazinon,  
20 Chlorpyrifos-methyl, Acephate, Prothiofos, Fosthiazate, Phosphocarb, Cadusafos and Dislufoton;

carbamate type compounds such as Carbaryl, Propoxur, Aldicarb, Carbofuran, Thiodicarb, Methomyl, Oxamyl, Ethiofencarb, Pirimicarb, Fenobucarb, Carbosulfan and  
25 Benfuracarb;

nelicetoxin derivatives such as Cartap and Thiocyclam;

organic chlorine type compounds such as Dicofol and Tetradifon;

organic metal type compounds such as Fenbutatin Oxide;

5       pyrethroid type compounds such as Fenvalerate, Permethrin, Cypermethrin, Deltamethrin, Cyhalothrin, Tefluthrin, Ethofenprox and Flufenprox;

benzoyl urea type compounds such as Diflubenzuron, Chlorfluazuron, Teflubenzuron and Flufenoxuron;

10       juvenile hormone-like compounds such as Methoprene;

pyridazinone type compounds such as Pyridaben;

pyrazole type compounds such as Fenpyroximate, Fipronil, Tebufenpyrad, Ethiprole, Tolefenpyrad and Acetoprole;

15       neonicotinoides such as Imidacloprid, Nitenpyram, Acetamiprid, Thiacloprid, Thiamethoxam, Clothianidin, Nidinetefuran and Dinotefuran;

hydrazine type compounds such as Tebufenozide, Methoxyfenozide and Chromafenozide;

20       pyridine type compounds such as Pyridaryl and Flonicamid;

tetronic acid type compounds such as Spirodiclofen;

strobilurin type compounds such as Fluacrypyrin;

dinitro type compounds, organosulfur compounds, urea  
25   type compounds, triazine type compounds, hydrozone type compounds and other compounds such as Buprofezin, Hexythiazox, Amitraz, Chlordimeform, Silafluofen,

Triazamate, Pymetrozine, Pyrimidifen, Chlorfenapyr, Indoxacarb, Acequinocyl, Etoxazole, Cyromazine and 1,3-dichloropropene; AKD-1022 and IKA-2000. Further, the fungicide of the present invention may also be mixed or  
5 used together with a microbial pesticide such as a BT agent or an insect pathogenic virus agent or an antibiotic such as Avermectin, Milbemycin, Spinosad or Emamectin Benzoate.

Of these other agricultural chemicals, examples of  
10 the active ingredient compounds of the fungicides (generic name; including compounds which are under application) include pyrimidinamine type compounds such as Mepanipyrim, Pyrimethanil and Cyprodinil, pyridinamine type compound such as Fluazinam;

15 azole type compounds such as Triadimefon, Bitertanol, Triflumizole, Etaconazole, Propiconazole, Penconazole, Flusilazole, Myclobutanil, Cyproconazole, Terbuconazole, Hexaconazole, Furconazole-cis, Prochloraz, Metconazole, Epoxiconazole, Tetraconazole, Oxpoconazole  
20 fumarate and Sipconazole;

quinoxaline type compounds such as Quinomethionate;  
dithiocarbamate type compounds such as Maneb, Zineb, Mancozeb, Polycarbamate, Metiram and Propineb;

organic chlorine type compounds such as Fthalide,  
25 Chlorothalonil and Quintozene;

imidazole type compounds such as Benomyl,  
Thiophanate-Methyl, Carbendazim and Cyazofamid;

- cyanoacetamide type compounds such as Cymoxanil;  
phenylamide type compounds such as Metalaxyl,  
Metalaxyl M, Oxadixyl, Ofurace, Benalaxyl, Furalaxyl and  
Cyprofuram;
- 5        sulfenic acid type compounds such as Dichlofluanid;  
copper type compounds such as Cupric hydroxide and  
Oxine Copper;
- isoxazole type compounds such as Hydroxyisoxazole;  
organophosphorus compounds such as Fosetyl-Al,
- 10    Tolcofos-Methyl, S-benzyl O, O-  
diisopropylphosphorothioate, O-ethyl S,S-  
diphenylphosphorodithioate and aluminum ethyl hydrogen  
phosphonate;
- N-halogenothioalkyl type compounds such as Captan,
- 15    Captafol and Folpet;
- dicarboxyimide type compounds such as Procymidone,  
Iprodione and Vinclozolin;
- benzanilide type compounds such as Flutolanil,  
Mepronil and Zoxamid;
- 20    piperazine type compounds such as Triforine;  
pyridine type compounds such as Pyrifenox;  
carbionol type compounds such as Fenarimol and  
Flutriafol;
- piperidine type compounds such as Fenpropidine;
- 25    morpholine type compounds such as Fenpropimorph;  
organotin type compounds such as Fentin Hydroxide  
and Fentin Acetate;

- urea type compounds such as Pencycuron;  
cinnamic acid type compounds such as Dimethomorph;  
phenyl carbamate type compounds such as  
Diethofencarb;
- 5       cyanopyrrole type compounds such as Fludioxonil and  
Fenpiclonil;  
      strobilurin type compounds such as Azoxystrobin,  
Kresoxim-Methyl, Metominofen, Triflouxystrobin,  
Picoxystrobin and Pyraclostrobin: (BAS 500F);
- 10      oxazolidinone type compounds such as Famoxadone; thiazole  
carboxamide type compounds such as Ethaboxam;  
      silyl amide type compounds such as Silthiopham;  
aminoacid amidecarbamate type compounds such as  
Iprovalicarb and Benthiavalicarb; Imidazolidine type
- 15      compounds such as fenamidone; hydroxyanilide type  
compounds such as Fenhexamid; benzene sulfonamide type  
compounds such as Flusulfamid; oxime ether type compounds  
such as Cyflufenamid; phenoxyamide type compounds such as  
Fenoxanil; triazole type compounds such as Simeconazole;
- 20      anthraquinone type compounds; crotonic acid type  
compounds; antibiotics and other compounds such as  
Isoprothiolane, Tricyclazole, Pyroquilon, Diclomezine,  
Pro. benazole, Quinoxifen, Propamocarb Hydrochloride,  
Spiroxamine, Chloropicrin, Dazomet and Metam-sodium; and
- 25      BJL-993, B JL-994, BAS-510, BAS-505, MTF-753 and UIBF-307.

Now, Test Examples of the agricultural and  
horticultural fungicides of the present invention will be

described below. However, the present invention is by no means restricted thereto. In each test, the controlling index was determined on the basis of the following standards.

5 [Controlling index]:[Degree of disease outbreak:Visual observation]

5 : No lesions nor sporogony recognizable

4 : Area of lesions, number of lesions or area of sporogony is less than 10% of non-treated plot

10 3 : Area of lesions, number of lesions or area of sporogony is less than 40% of non-treated plot

2 : Area of lesions, number of lesions or area of sporogony is less than 70% of non-treated plot

15 1 : Area of lesions, number of lesions or area of sporogony is at least 70% of non-treated plot

#### TEST EXAMPLE 1

#### Tests on preventive effect against wheat powdery mildew

Wheat (cultivar: Norin-61-go) was cultivated in a polyethylene pot having a diameter of 7.5 cm, and when  
20 the wheat reached a one and a half-leaf stage, the wheat was sprayed with 10 ml of a drug solution having a predetermined concentration of the compound of the present invention by a spray gun. After the drug solution dried, the wheat was inoculated by spreading  
25 with conidiospore of fungi of powdery mildew, and the wheat was kept in a thermostatic chamber at 20°C. From 6 to 8 days after the inoculation, the area of sporogony

was examined to determine the controlling index in accordance with the above evaluation standards. As a result, of the above compounds, compounds Nos. 1, 2, 8, 47, 58, 61, 62, 69, 73, 76, 77, 78, 83, 87, 91, 107, 110, 112, 114, 117, 119, 138, 250, 262 and 274 showed effects with a controlling index of 4 or above at a concentration of 500 ppm, and the compounds Nos. 3, 4, 5, 6, 7, 9, 10, 11, 13, 14, 18, 19, 23, 27, 30, 31, 32, 33, 34, 35, 36, 38, 40, 41, 43, 50, 51, 54, 55, 56, 59, 65, 72, 74, 75, 82, 84, 89, 90, 92, 93, 94, 99, 100, 101, 102, 103, 104, 105, 106, 108, 109, 111, 113, 118, 120, 121, 122, 123, 124, 133, 136, 142, 186, 187, 188, 189, 190, 191, 192, 193, 194, 199, 200, 210, 211, 213, 228, 243, 245, 249, 252, 254, 272, 287, 288, 289, 290, 291 and 292 showed effects with a controlling index of 4 or above at a concentration of 125 ppm.

#### TEST EXAMPLE 2

##### Test on preventive effect against rice blast

Rice (cultivar: Nihonbare) was calculated in a polyethylene pot having a diameter of 7.5 cm, and when the rice reached a one and a half-leaf stage, the rice was sprayed with 10 ml of a drug solution having a predetermined concentration of the compound of the present invention by a spray gun. After the drug solution dried, the rice was sprayed and inoculated with a conidiospore suspension of fungi of rice blast, and the rice was kept in an inoculation box at 20°C for 24 hours,

and then kept in a thermostatic chamber at 20°C. From 6 to 11 days after the inoculation, the number of lesions was examined to determine the controlling index in accordance with the above evaluation standards. As a result, of the above compounds, the compounds Nos. 31, 56, 76, 90, 103 and 136 showed effects with a controlling index of 4 or above at a concentration of 500 ppm, and the compounds Nos. 50, 74, 75 and 102 showed effects with a controlling index of 4 or above at a concentration of 125 ppm.

#### TEST EXAMPLE 3

##### Test on preventive effect against eggplant powdery mildew

Eggplant (cultivar: Senryo-2-go) was cultivated in a polyethylene pot having a diameter of 7.5 cm, and when the eggplant reached a two-leaf stage, the eggplant was sprayed with 10 ml of a drug solution having a predetermined concentration of the compound of the present invention by a spray gun. After the drug solution dried, the eggplant was inoculated by spreading with conidiospore of fungi of eggplant powdery mildew, and the eggplant was kept in a thermostatic chamber at 20°C. 16 days after the inoculation, the area of sporogony was examined to determine the controlling index in accordance with the above evaluation standards. As a result, of the above compounds, compounds Nos. 1, 3, 5, 7, 92, 101 and 103 showed effects with a controlling index of 4 or above at a concentration of 500 ppm, and

the compounds Nos. 9, 11, 55, 90 and 102 showed effects with a controlling index of 4 or above at a concentration of 125 ppm.

#### TEST EXAMPLE 4

##### 5 Test on preventive effect against cucumber powdery mildew

Cucumber (cultivar: Suvo) was cultivated in a polyethylene pot having a diameter of 7.5 cm, and when the cucumber reached one and a half-leaf stage, the cucumber was sprayed with a 10 ml of a drug solution  
10 having a predetermined concentration of the compound of the present invention by a spray gun. After the drug solution dried, the cucumber was sprayed and inoculated with a conidiospore suspension of fungi of powdery mildew, and the cucumber was kept in a thermostatic  
15 chamber at 20 °C. From 7 to 11 days after the inoculation, the area of sporogony was examined to determine the controlling index in accordance with the above evaluation standards. As a result, of the above compounds, the compound No. 98 showed effects with a  
20 controlling index of 4 or above at a concentration of 500 ppm, and compounds Nos. 1, 5, 7, 9, 55, 74, 90, 92, 93, 102, 103, 123 and 124 showed effects with a controlling index of 4 or above at a concentration of 125 ppm.

Now, Formulation Examples of the compounds of the  
25 present invention will be described below. However, the formulation dose, the dosage form or the like is by no means restricted to the following Examples.

## FORMULATION EXAMPLE 1

- |   |                                       |                    |
|---|---------------------------------------|--------------------|
|   | (1) Compound of the present invention | 20 parts by weight |
|   | (2) Clay                              | 72 parts by weight |
| 5 | (3) Sodium lignin sulfonate           | 8 parts by weight  |

The above components are uniformly mixed to obtain a wettable powder.

## FORMULATION EXAMPLE 2

- |    |                                       |                    |
|----|---------------------------------------|--------------------|
|    | (1) Compound of the present invention |                    |
| 10 |                                       | 5 parts by weight  |
|    | (2) Talc                              | 95 parts by weight |

The above components are uniformly mixed to obtain a dust.

## FORMULATION EXAMPLE 3

- |    |  |                    |
|----|--|--------------------|
| 15 | (1) Compound of the present invention  |                    |
|    |  | 20 parts by weight |
|    | (2) N,N'-dimethylacetamide             | 20 parts by weight |
|    | (3) Polyoxyethylene alkyl phenyl ether |                    |
|    |  | 10 parts by weight |
| 20 | (4) Xylene                             | 50 parts by weight |

The above components are uniformly mixed and dissolved to obtain an emulsifiable concentrate.

## FORMULATION EXAMPLE 4

- |    |  |                    |
|----|--|--------------------|
|    | (1) Clay                               | 68 parts by weight |
| 25 | (2) Sodium lignin sulfonate            | 2 parts by weight  |
|    | (3) Polyoxyethylene alkyl aryl sulfate |                    |
|    |  | 5 parts by weight  |

100

(4) Fine silica 25 parts by weight

A mixture of the above components and the compound of the present invention are mixed in a weight ratio of 4:1 to obtain a wettable powder.

5 FORMULATION EXAMPLE 5

(1) Compound of the present invention

50 parts by weight

(2) Oxyated polyalkylphenyl phosphate-

triethanolamine

2 parts by weight

10

(3) Silicone

0.2 part by weight

(4) Water

47.8 parts by weight

The above components are uniformly mixed and pulverized to obtain a stock solution, and

(5) Sodium polycarboxylate

5 parts by weight

15

(6) Anhydrous sodium sulfate

42.8 parts by weight

are further added thereto, followed by uniform mixing, granulation and drying to obtain a granular wettable powder.

FORMULATION EXAMPLE 6

20

(1) Compound of the present invention

5 parts by weight

(2) Polyoxyethylene octylphenyl ether

1 part by weight

(3) Phosphate of polyoxyethylene

25

0.1 part by weight

(4) Particulate calcium carbonate

93.9 parts by weight

The above components (1) to (3) are preliminarily mixed uniformly and diluted with a proper amount of acetone, the diluted mixture is sprayed on the component (4), and acetone is removed to obtain granules.

5 FORMULATION EXAMPLE 7

- |                                       |                      |
|---------------------------------------|----------------------|
| (1) Compound of the present invention | 2.5 parts by weight  |
| (2) N-methyl-2-pyrrolidone            | 2.5 parts by weight  |
| (3) Soybean oil                       | 95.0 parts by weight |

10 The above components are uniformly mixed and dissolved to obtain an ultra low volume formulation.

FORMULATION EXAMPLE 8

- |  |                      |
|--|----------------------|
| (1) Compound of the present invention                    | 20 parts by weight   |
| 15 (2) Oxyated polyalkylphenol phosphate triethanolamine | 2 parts by weight    |
| (3) Silicone   | 0.2 part by weight   |
| (4) Xanthan gum  | 0.1 part by weight   |
| (5) Ethylene glycol                                      | 5 parts by weight    |
| 20 (6) Water   | 72.7 parts by weight |

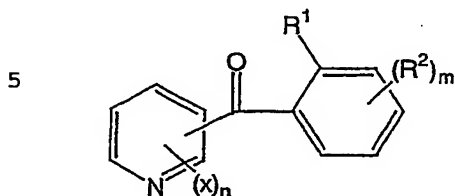
The above components are uniformly mixed and pulverized to obtain an aqueous suspension.

INDUSTRIAL APPLICABILITY

As mentioned above, the benzoylpyridine derivative  
25 represented by the formula (I) or its salt has excellent effects as an active ingredient of a fungicide.

CLAIMS

1. A benzoylpyridine derivative represented by the formula (I) or its salt:

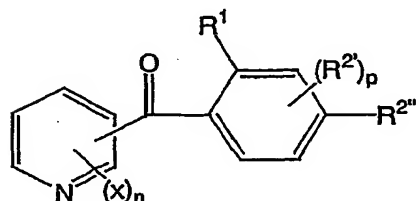


wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R¹ is a substitutable alkyl group; R² is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group; and m is 1, 2, 3 or 4, provided that when m is at least 2, R² may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and n is 1, m is 1 or 2).

25 2. The benzoylpyridine derivative or its salt according to Claim 1, wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group,

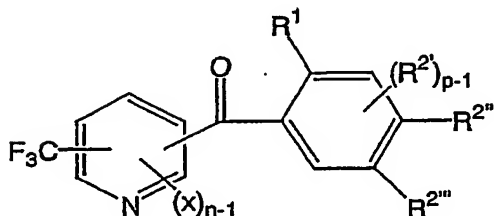
a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group or a substitutable amino group.

- 5 3. The benzoylpyridine derivative or its salt according to Claim 1, wherein m is 2, 3 or 4.
4. The benzoylpyridine derivative or its salt according to Claim 1, wherein  $R^1$  is an alkyl group, and  $R^2$  is an alkyl group, an alkoxy group or a hydroxyl group.
- 10 5. The benzoylpyridine derivative or its salt according to Claim 1, which is represented by the formula (I'):



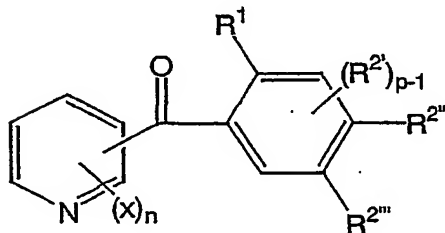
- wherein X, n and  $R^1$  are as defined in Claim 1,  $R^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group, p is 1, 2 or 3, and  $R^{2''}$  is a substitutable alkoxy group or a hydroxyl group, provided that at least two of  $R^{2'}$  and  $R^{2''}$  may contain an oxygen atom to form a condensed ring
- 15
  - 20 (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and n is 1, p is 1).

6. The benzoylpyridine derivative or its salt according to Claim 5, which is represented by the formula (I''):



wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group or a substitutable cycloalkoxy group, p is 1, 2 or 3, and each of R<sup>2''</sup> and R<sup>2'''</sup> is a substitutable alkoxy group.

7. The benzoylpyridine derivative or its salt according to Claim 5, which is represented by the formula (I'''):

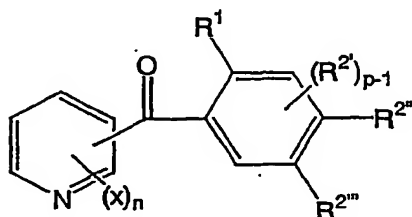


wherein X is a halogen atom, a substitutable alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; n is 1,

105

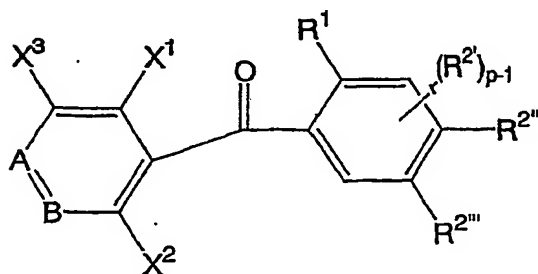
2, 3 or 4;  $R^1$  is an alkyl group;  $R^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group or a substitutable cycloalkoxy group;  $p$  is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is a substitutable alkoxy group.

- 5 8. The benzoylpyridine derivative or its salt according to Claim 5, which is represented by the formula (I'''):



wherein  $X$  is a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $n$  is 1, 2 or 3;  
 10  $R^1$  is an alkyl group;  $R^{2'}$  is an alkoxy group;  $p$  is 1, 2 or 3, and each of  $R^{2''}$  and  $R^{2'''}$  is an alkoxy group.

9. The benzoylpyridine derivative or its salt according to Claim 8, which is represented by the formula (I'''):



15

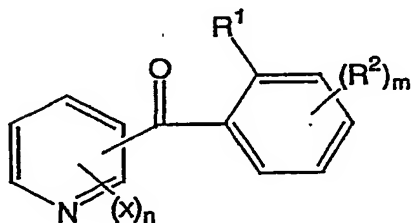
wherein  $B$  is  $-CX^4=$  when  $A$  is  $-N=$ ;  $B$  is  $-N=$  when  $A$  is  $-CH=$ ; each of  $X^1$  and  $X^2$  which are independent of each other, is a halogen atom, an alkoxy group, an alkyl group,

a CF<sub>3</sub> group or an alkylthio group; X<sup>3</sup> is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; X<sup>4</sup> is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a CF<sub>3</sub> group or an alkylthio group; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is an alkoxy group; p is 1, 2 or 3; and each of R<sup>2''</sup> and R<sup>2'''</sup> is an alkoxy group.

10. A fungicide which contains the benzoylpyridine derivative represented by the formula (I) or its salt as defined in Claim 1 as an active ingredient.

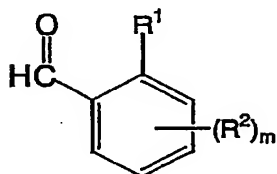
11. A fungicide which contains the benzoylpyridine derivative represented by the formula (I') or its salt as defined in Claim 5 as an active ingredient.

12. A process for producing a benzoylpyridine derivative represented by the formula (I) or its salt:

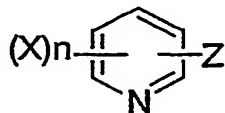


{wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is a substitutable alkyl

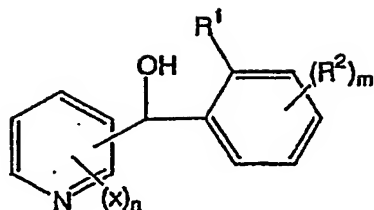
group;  $R^2$  is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group; and  $m$  is 1, 2, 3 or 4, provided that when  $m$  is at least 2,  $R^2$  may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and  $n$  is 1,  $m$  is 1 or 2)), which process comprises reacting a substituted benzaldehyde represented by the formula (VI-1):



(wherein  $R^1$ ,  $R^2$  and  $m$  are as defined above) and a metal salt of a substituted pyridine derivative represented by the formula (VII-1):

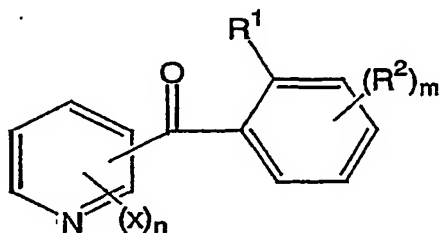


(wherein  $X$  is as defined above, and  $Z$  is a metal atom or a composite salt thereof) to produce phenylpyridyl methanol represented by the formula (X):



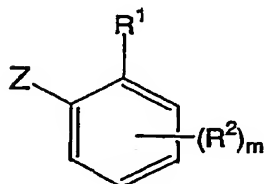
(wherein X, n, m, R<sup>1</sup> and R<sup>2</sup> are as defined above, with a proviso as in formula (I)), and oxidizing it.

13. A process for producing a benzoylpyridine derivative  
5 represented by the formula (I) or its salt:

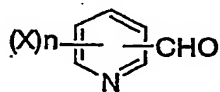


{wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a  
10 substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is a substitutable alkyl group; R<sup>2</sup> is a substitutable alkyl group, a substitutable  
15 alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group; and m is 1, 2, 3 or 4, provided that when m is at least 2, R<sup>2</sup> may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted

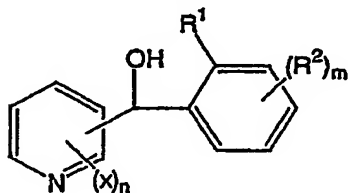
by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and n is 1, m is 1 or 2)), which process comprises reacting a metal salt of a substituted benzene derivative represented by the formula (VI-2):



(wherein R¹, R² and m are as defined above, and Z is a metal atom or a composite salt thereof) and a substituted pyridyl aldehyde represented by the formula (VII-2):



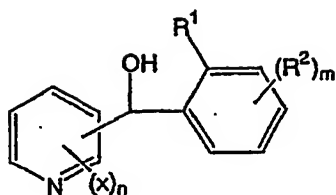
(wherein X is as defined above) to produce phenylpyridyl methanol represented by the formula (X):



(wherein X, n, m, R¹ and R² are as defined above with a proviso as in formula (I)), and oxidizing it.

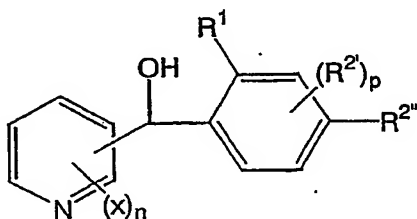
14. Phenylpyridyl methanol represented by the formula (X):

110



wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a hydroxyl group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is a substitutable alkyl group; R<sup>2</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group; and m is 1, 2, 3 or 4, provided that when m is at least 2, R<sup>2</sup> may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and n is 1, m is 1 or 2).

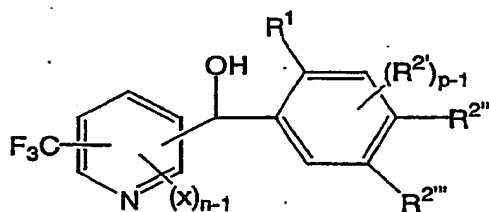
15. The phenylpyridyl methanol according to Claim 14, which is represented by the formula (X''):



111

wherein X, n and R<sup>1</sup> are as defined in Claim 14, R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group or a hydroxyl group, p is 1, 2 or 3, and R<sup>2''</sup> is a substitutable alkoxy group or a hydroxyl group, provided that at least two of R<sup>2'</sup> and R<sup>2''</sup> may contain an oxygen atom to form a condensed ring (excluding a case where the pyridine ring is substituted by a benzoyl group at the 2-position; the pyridine ring is substituted by an alkoxy group, a hydroxyl group or a benzyloxy group at the 3-position; and n is 1, p is 1).

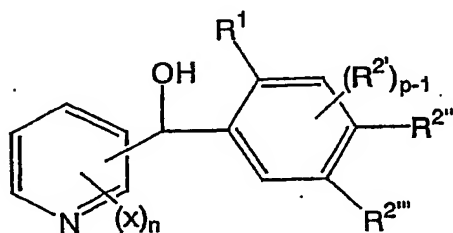
16. The phenylpyridyl methanol according to Claim 15, which is represented by the formula (X''):



wherein X is a halogen atom, a nitro group, a substitutable alkoxy group, a substitutable aryloxy group, a substitutable cycloalkoxy group, a substitutable hydrocarbon group, a substitutable alkylthio group, a cyano group, a carboxyl group which may be esterified or amidated, or a substitutable amino group; n is 1, 2, 3 or 4; R<sup>1</sup> is an alkyl group; R<sup>2'</sup> is a substitutable alkyl group, a substitutable alkoxy group, a substitutable aryloxy group or a substitutable cycloalkoxy group, p is

1, 2 or 3, and each of  $R^{2''}$  and  $R^{2'''}$  is a substitutable alkoxy group.

17. The phenylpyridyl methanol according to Claim 15, which is represented by the formula ( $X'''$ ):

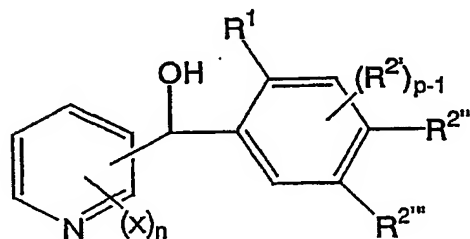


5

wherein X is a halogen atom, a substitutable alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group; n is 1, 2, 3 or 4;  $R^1$  is an alkyl group;  $R^{2'}$  is a substitutable alkyl group, a substitutable alkoxy group or a substitutable cycloalkoxy group; p is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is a substitutable alkoxy group.

10

18. The phenylpyridyl methanol according to Claim 15, which is represented by the formula ( $X''''$ ):



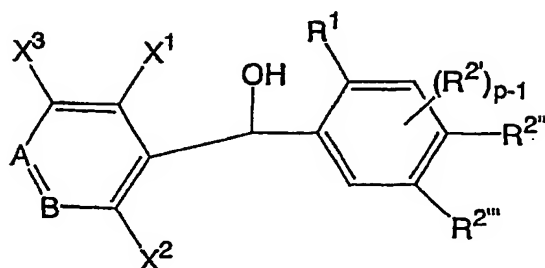
15

wherein X is a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group; n is 1, 2 or 3;  $R^1$  is an alkyl group;  $R^{2'}$  is an alkoxy group; p is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is an alkoxy group.

19. The phenylpyridyl methanol according to Claim 18,

113

which is represented by the formula (X'''''):



wherein B is  $-CX^4=$  when A is  $-N=$ ; B is  $-N=$  when A is  $-CH=$ ; each of  $X^1$  and  $X^2$  which are independent of each other, is a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $X^3$  is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $X^4$  is a hydrogen atom, a halogen atom, an alkoxy group, an alkyl group, a  $CF_3$  group or an alkylthio group;  $R^1$  is an alkyl group;  $R^{2'}$  is an alkoxy group; p is 1, 2 or 3; and each of  $R^{2''}$  and  $R^{2'''}$  is an alkoxy group.

**THIS PAGE BLANK (USPTO)**

## INTERNATIONAL SEARCH REPORT

II National Application No

PCI/JP 01/05851

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C07D213/61 C07D213/64 C07D213/80 C07D213/70 C07D213/74  
C07D213/26 A01N43/40

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C07D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data, BIOSIS, CHEM ABS Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 00 15616 A (DOW AGROSCIENCES LLC) 23 March 2000 (2000-03-23) page 23; example 6	1-4, 10
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; ARNOLDI, ANNA ET AL: "Synthesis of some 3-phenyl-1-substituted(or 1,1-disubstituted)prop-2-yn-1-ols and their in vivo activity against some phytopathogenic fungi" retrieved from STN Database accession no. 99:153745 CA XP002180437 abstract; RN 87446-23-9 & PESTIC. SCI. (1982), 13(6), 670-8 , 1982,	1-4, 10

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

\*A\* document defining the general state of the art which is not considered to be of particular relevance

\*E\* earlier document but published on or after the international filing date

\*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

\*O\* document referring to an oral disclosure, use, exhibition or other means

\*P\* document published prior to the international filing date but later than the priority date claimed

\*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

\*&amp;\* document member of the same patent family

Date of the actual completion of the international search

19 October 2001

Date of mailing of the international search report

13/11/2001

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Frelon, D

## INTERNATIONAL SEARCH REPORT

In International Application No

PCT/JP 01/05851

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 94 24106 A (ZENECA LTD) 27 October 1994 (1994-10-27) examples 2-4,7,8,13-17,19 ---	14
X	WO 98 06700 A (BANYU PHARMA CO LTD ;KATO YOSHIKI (JP); TILLYER RICHARD D (US); D) 19 February 1998 (1998-02-19) page 36; example 28 ---	1-3
X	WO 99 41237 A (CONNOLLY DANIEL T ;SCHILLING ROGER J (US); GLENN KEVIN C (US); LEE) 19 August 1999 (1999-08-19) cited in the application examples 263,284,288,290,292,293; 266,273,275,296; tables 9,10,11,12 ---	1,14
X	EP 0 032 516 A (LACER SA) 29 July 1981 (1981-07-29) examples 2a,5,9,15,19,20,23 ---	1-4,14
X	EP 0 177 054 A (RICHTER GEDEON VEGYESZET) 9 April 1986 (1986-04-09) example 9 ---	14
X	EP 0 177 907 A (RICHTER GEDEON VEGYESZET) 16 April 1986 (1986-04-16) example 4 ---	1-4
X	EP 0 704 435 A (EISAI CO LTD ;EISAI KAGAKU KK (JP)) 3 April 1996 (1996-04-03) examples 9,10 ---	14
X	EP 0 848 000 A (TANABE SEIYAKU CO) 17 June 1998 (1998-06-17) Reference examples 1,3,12,15 ---	1-3
X,P	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 21, 3 August 2001 (2001-08-03) & JP 2001 089412 A (OTSUKA PHARMACEUT CO LTD), 3 April 2001 (2001-04-03) abstract -& DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; XP002180606 Retrieved from STN Database accession no. 134:266205 abstract; RN 332010-49-8 ---	1
	--- -/--	

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/JP 01/05851

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 05, 14 September 2000 (2000-09-14) & JP 2000 063275 A (TANABE SEIYAKU CO LTD), 29 February 2000 (2000-02-29) abstract -& DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; XP002180607 Retrieved from STN Database accession no. 132:194292 abstract; RNs 209262-21-5, 209262-23-7, 209262-49-7, 259817 -61-3, 209262-50-0 ---	1
X	PATENT ABSTRACTS OF JAPAN vol. 1996, no. 03, 29 March 1996 (1996-03-29) -& JP 07 309837 A (TAKEDA CHEM IND LTD), 28 November 1995 (1995-11-28) cited in the application compound A18, page 38; compounds C1, C5, C11, C12, page 43; compound D10, page 45; compound E17, page 47; compounds E27, E28, E31, page 48 ---	1, 14
X	SUGAHARA, MASAKATSU ET AL: "A synthesis of 1-pyridyl-naphthalene lignan analogs" TETRAHEDRON LETT. (1998), 39(11), 1377-1380, 1998, XP002180435 substrates 4f, 4g, 4h ---	14, 15
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; MATAS DOCAMPO, RICARDO ET AL: "2(1H)-Pyridinones" retrieved from STN Database accession no. 98:107169 CA XP002180438 abstract; RNs 80099-82-7, 80099-92-9, 80100-03-4, 80100-07- 8, 80100-10-3, 80100-54-5, 84852-99-3, 84853-0 1-0, 84853-17-8, 84853-59-8, 84853-62-3, 84865 -20-3, 84854-04-6, 84854-05-7, 84853-18-9, 848 53-23-6, etc & ES 506 367 A (LACER S. A., SPAIN) 16 August 1982 (1982-08-16) ---	1-4, 14
A	WO 96 17829 A (ZENECA LTD ; KANNE DAVID B (US)) 13 June 1996 (1996-06-13) cited in the application page 3; claims -----	1-18

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No  
PCT/JP 01/05851

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 0015616	A	23-03-2000	AU 6049199 A WO 0015616 A1	03-04-2000 23-03-2000
WO 9424106	A	27-10-1994	US 5308826 A AT 163408 T AU 676732 B2 AU 6542394 A BR 9406570 A CA 2161115 A1 CN 1124024 A CZ 9502705 A3 DE 69408663 D1 DK 695292 T3 EG 20346 A EP 0695292 A1 ES 2112536 T3 FI 955012 A WO 9424106 A1 GR 3026239 T3 HU 73747 A2 JP 9500095 T MX 9402950 A1 NO 954214 A NZ 265087 A PL 311226 A1 SK 131295 A3 US 5585327 A ZA 9402769 A ZW 5294 A1	03-05-1994 15-03-1998 20-03-1997 08-11-1994 06-02-1996 27-10-1994 05-06-1996 15-05-1996 02-04-1998 23-09-1998 31-10-1998 07-02-1996 01-04-1998 22-11-1995 27-10-1994 29-05-1998 30-09-1996 07-01-1997 31-01-1995 20-12-1995 24-06-1997 05-02-1996 05-06-1996 17-12-1996 26-01-1995 10-08-1994
WO 9806700	A	19-02-1998	AU 711936 B2 AU 3975797 A EP 0923557 A1 JP 11514676 T WO 9806700 A1 US 5962688 A	28-10-1999 06-03-1998 23-06-1999 14-12-1999 19-02-1998 05-10-1999
WO 9941237	A	19-08-1999	AU 3285499 A WO 9941237 A1	30-08-1999 19-08-1999
EP 0032516	A	29-07-1981	EP 0032516 A1 AT 7295 T DE 3067662 D1 ES 498507 D0 ES 8206480 A1 ES 498508 D0 ES 8200343 A1 ES 498509 D0 ES 8200344 A1 ES 498510 D0 ES 8200345 A1 JP 56120668 A PT 72354 A , B US 4614833 A US 4736037 A	29-07-1981 15-05-1984 07-06-1984 01-08-1982 16-11-1982 16-11-1981 16-01-1982 16-11-1981 16-01-1982 16-11-1981 16-01-1982 22-09-1981 01-02-1981 30-09-1986 05-04-1988
EP 0177054	A	09-04-1986	HU 38908 A2 AT 40687 T	28-07-1986 15-02-1989

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/JP 01/05851

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 0177054	A		AU 580096 B2	22-12-1988
			AU 4833085 A	10-04-1986
			CN 85107279 A	17-09-1986
			DD 238230 A5	13-08-1986
			DE 3568164 D1	16-03-1989
			DK 455185 A	06-04-1986
			EP 0177054 A1	09-04-1986
			ES 547603 D0	01-05-1987
			ES 8705391 A1	16-07-1987
			ES 554310 D0	01-07-1987
			ES 8706636 A1	16-09-1987
			ES 554311 D0	01-07-1987
			ES 8706637 A1	16-09-1987
			FI 853868 A	06-04-1986
			GR 852408 A1	04-02-1986
			IL 76578 A	31-03-1989
			IN 162569 A1	11-06-1988
			JP 61091173 A	09-05-1986
			NO 853937 A	07-04-1986
			PH 21375 A	15-10-1987
			PL 255642 A1	05-10-1987
			PT 81257 A ,B	01-11-1985
			US 4624959 A	25-11-1986
			US 4691017 A	01-09-1987
			YU 158285 A1	30-04-1988
			ZA 8507677 A	25-06-1986
EP 0177907	A	16-04-1986	HU 38907 A2	28-07-1986
			AT 34739 T	15-06-1988
			AU 579665 B2	01-12-1988
			AU 4832885 A	10-04-1986
			CN 85107284 A	10-06-1986
			DD 238384 A5	20-08-1986
			DE 3563048 D1	07-07-1988
			DK 455285 A	06-04-1986
			EP 0177907 A1	16-04-1986
			ES 547604 D0	01-04-1987
			ES 8704460 A1	16-06-1987
			FI 853866 A	06-04-1986
			GR 852409 A1	04-02-1986
			IN 162570 A1	11-06-1988
			JP 61091172 A	09-05-1986
			NO 853935 A	07-04-1986
			PH 21435 A	15-10-1987
			PL 255643 A1	12-01-1987
			PT 81255 A ,B	01-11-1985
			SU 1346042 A3	15-10-1987
			US 4713388 A	15-12-1987
			US 4691017 A	01-09-1987
			YU 158385 A1	30-04-1988
			ZA 8507676 A	25-06-1986
EP 0704435	A	03-04-1996	DE 69411273 D1	30-07-1998
			DE 69411273 T2	03-12-1998
			EP 0704435 A1	03-04-1996
			US 5637716 A	10-06-1997
			WO 9504044 A1	09-02-1995
			JP 8092214 A	09-04-1996

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/JP 01/05851

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 0848000	A	17-06-1998	CA 2224635 A1	13-06-1998
			CN 1184813 A	17-06-1998
			EP 0848000 A1	17-06-1998
			JP 10226685 A	25-08-1998
			SG 53130 A1	28-09-1998
			US 5965730 A	12-10-1999
JP 2001089412	A	03-04-2001	NONE	
JP 2000063275	A	29-02-2000	NONE	
JP 07309837 5	A		NONE	
ES 506367	A		NONE	
WO 9617829	A	13-06-1996	US 5565413 A	15-10-1996
			AU 4509696 A	26-06-1996
			BG 101557 A	31-08-1998
			BR 9509951 A	14-10-1997
			CA 2206740 A1	13-06-1996
			CN 1168666 A	24-12-1997
			CZ 9701663 A3	17-09-1997
			EP 0796246 A1	24-09-1997
			HU 77881 A2	28-09-1998
			JP 11500410 T	12-01-1999
			PL 320536 A1	13-10-1997
			SK 70597 A3	08-10-1997
			WO 9617829 A2	13-06-1996